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LWR Fuel: Performance and research challenges

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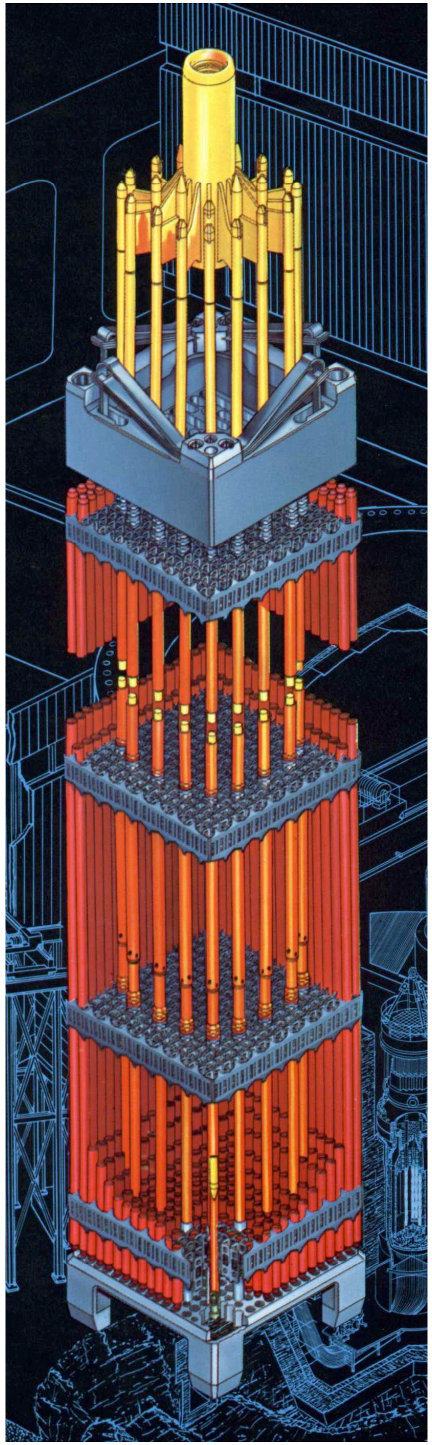


Contents

- Fundamental requirements of fuel
- A few research challenges
- Materials for fuel and cladding
- Fuel manufacture
- What happens to fuel in a reactor

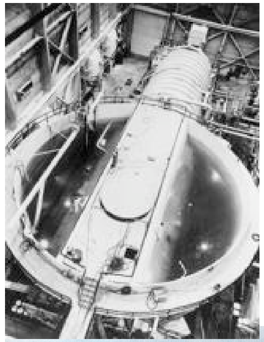
Important definition

= football ✗

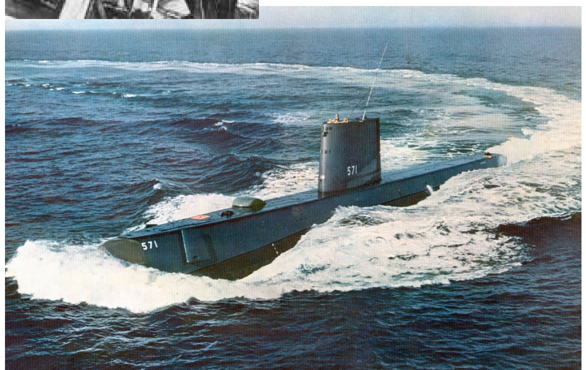
= football ✓

Why research LWR fuel?

LWRs have been around for > 50 years: the fuel must be well understood by now!



SW1, 1953



USS Nautilus, 1954

Shippingport, 1957

Yes, but ….

- LWR systems likely to be around for at least another 60+ years
- Fuel is the only major component that we plan on changing regularly
- Fuel duties today bear little resemblance to those of 50 years ago
- Continuous efforts to improve economics and safety
- The future may bring new priorities: transmutation, recycle, safety, proliferation resistance, …

What do we want to achieve?

- Improved safety:
  - Avoid fuel failure during normal operation and frequent faults
  - Reduced fuel damage and degradation under accident conditions
- Improved fuel cycle economics
- Improved operational flexibility
- Improved sustainability

A few research challenges …

Avoid fuel failure during normal operation and frequent faults

Reduced fuel damage and degradation under accident conditions

- Understanding of failure and degradation mechanisms
- Identification and development of mitigating approaches
  - Improved cladding materials
  - Improved fuel materials

Improve fuel cycle economics

- Reduced manufacturing costs
- Simplified processes / simplified designs / reduced scrap
- Higher burnup (up to the economically optimal point)

Improve operational flexibility

- Facilitate load-follow and frequency-follow operation (tolerance to power manoeuvres)
- Longer cycle lengths
- Simplify leak detection

Improved sustainability

- Ability to burn Pu as MOX / transmutation of Pu (and Np)
- Reduced resource requirements
- Alternative fuel materials (e.g. thorium)
- Reduced environmental impact

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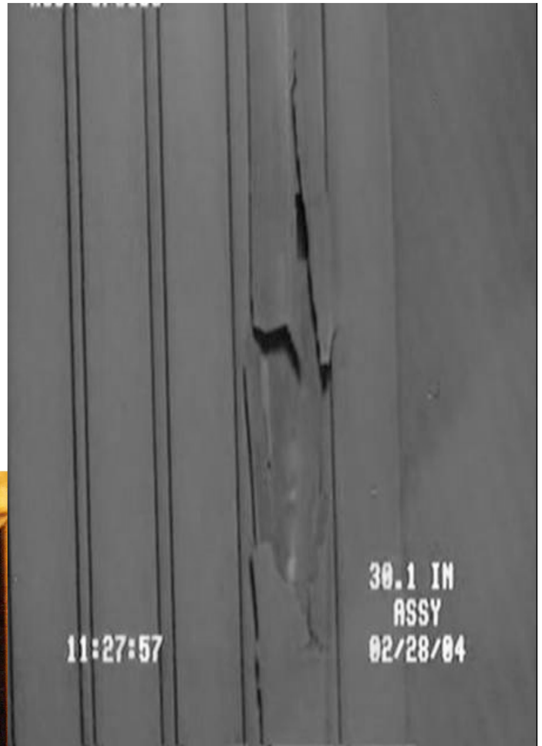
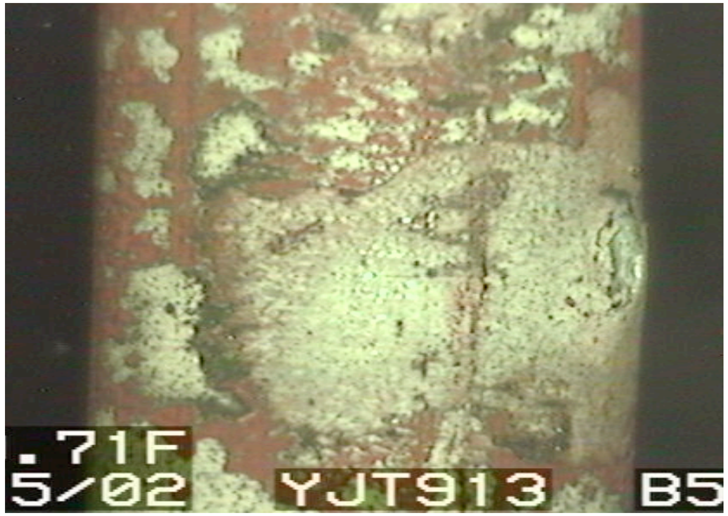
How does LWR fuel fail?

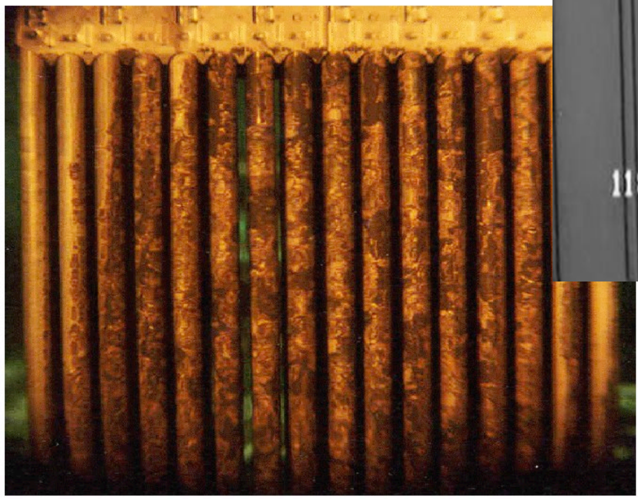
Grid Fretting

Secondary hydriding

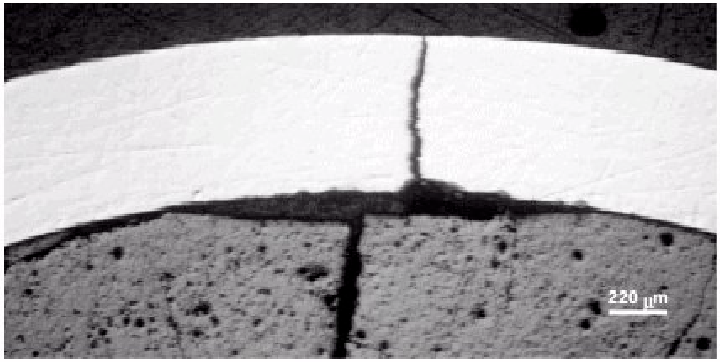


Corrosion





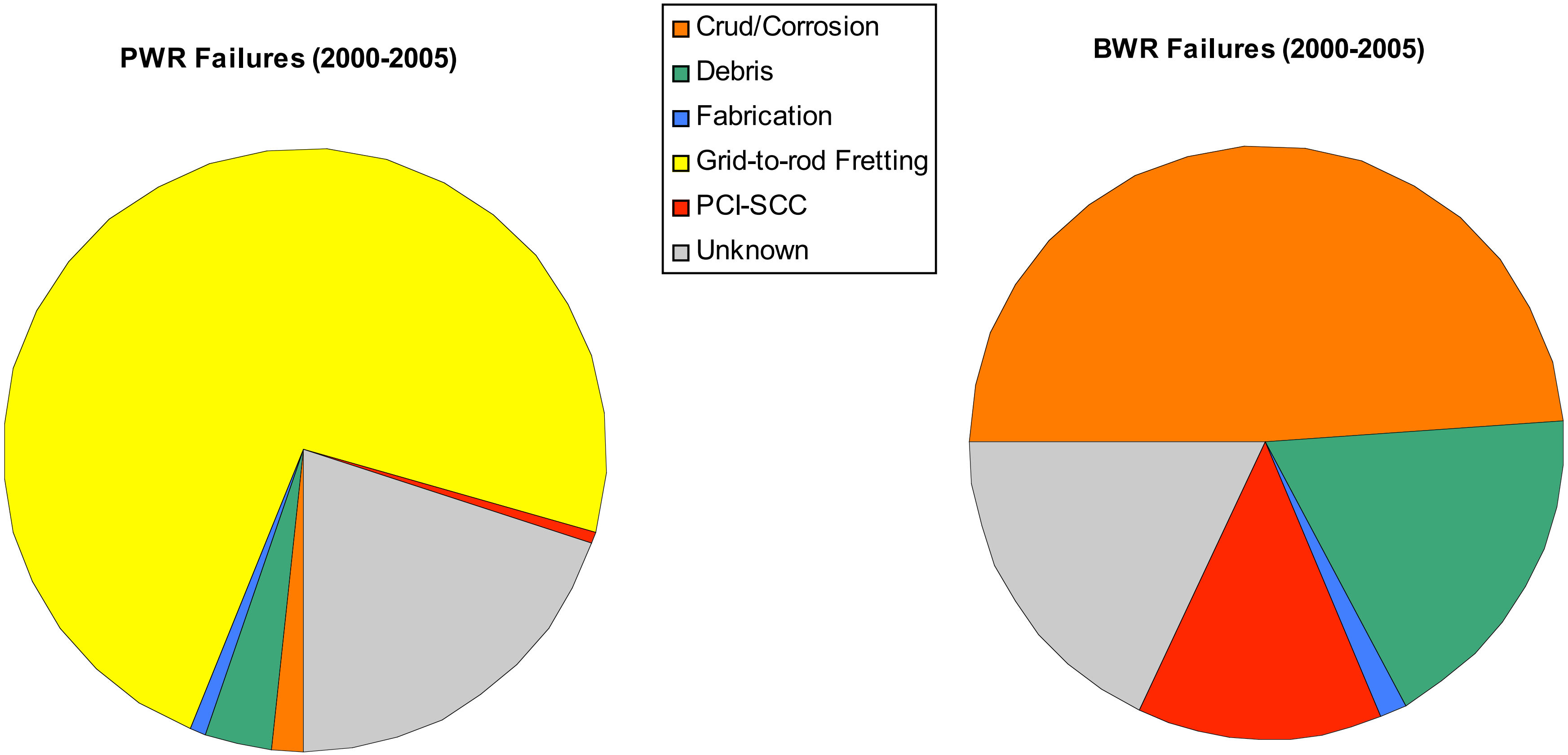
Crudding



Pellet –Clad Interaction (PCI)

Source: Westinghouse and EPRI

Fuel Failures By Mechanism



Source: Todd Allen / EPRI

A reminder of the basics ….

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Fuel Functional Requirements

- Provide the primary heat source
- Provide physical location for the fissile material
- Prevent relocation of the fuel into a more reactive configuration
- Protect fissile material from erosion and corrosion by the primary coolant
- Provide a reliable primary barrier to the release of radioactivity
- Provide a controlled path for the primary coolant and facilitate heat transport from the fissile material to the coolant
- Provide a compact structural unit that can be easily moved in and out of the core by a refueling machine

Design Objectives

- Safety — Must have low probability of failure during all anticipated operational conditions and the more frequent fault conditions. The consequences of any failure must be benign, i.e. the failure must not propagate. Failed fuel must retain structural integrity.
- Economy — Produce the required energy over the specified time at minimum cost.
- Reliability — Must support reliable and predictable plant operations (related to economy, above).
- Operations — Facilitate ease of operations, both whilst producing power and during handling.
- Function — Must satisfy specific functional requirements, such as load-follow, frequency-follow, etc.

Materials

Candidate fuel element materials

- Fuel materials
  - Oxides: UO_2, $(U,Pu)O_2$
  - Carbides: UC, (U,Pu)C
  - Nitrides: UN, (U,Pu)N
  - Metal Alloys: U-Pu-Zr
  - Others: UAl_x, U_3Si_2, U/Zr hydride, UCO, …
- Structural materials
  - Stainless steel
  - Inconel
- Cladding materials
  - Zirconium alloys
  - Stainless steels
  - Aluminium and/or magnesium alloys (research reactors, early gas reactors)
  - Refractory alloys (e.g. Ni, W, Nb, Mo, V, …)
  - Coatings (e.g.PyC, SiC, ZrC, …)
  - Composites (e.g. SiC-SiC)

Fuel Material

- Fuel material should :
  - Withstand peak operating temperature without melt or other degradation
  - Not deteriorate with burnup due to fission product build-up
  - Preferably retain the volatile fission products such as iodine, caesium

Clad Material

- Clad material should :
  - Be corrosion resistant under the prevailing conditions
  - Avoid melting or other degradation mechanisms
  - Not interact adversely with the fuel
  - Have a low cross-section for neutron absorptions
  - Not deteriorate excessively under neutron irradiation

Zirconium alloys are the universal choice for LWR applications

Some research challenges in Zr alloy cladding technology

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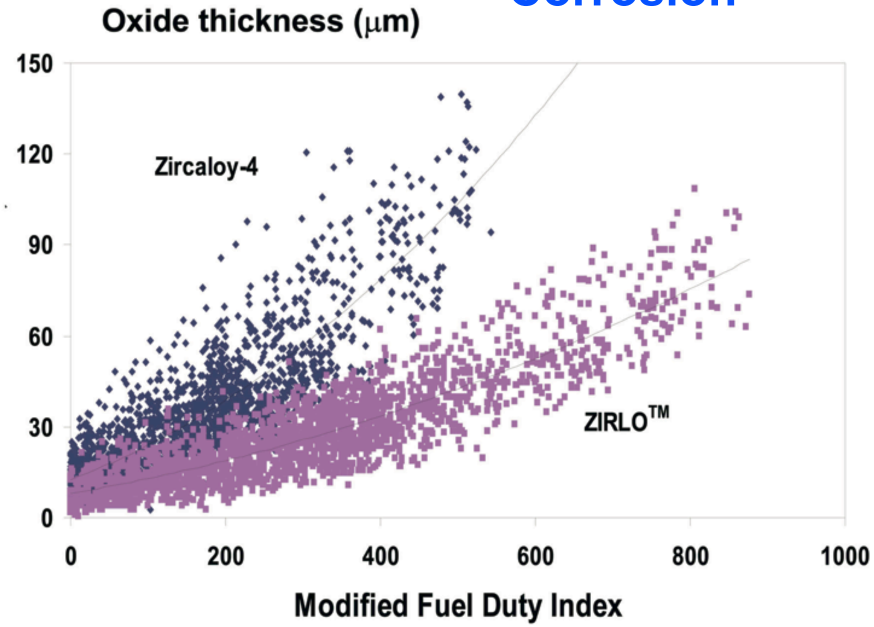
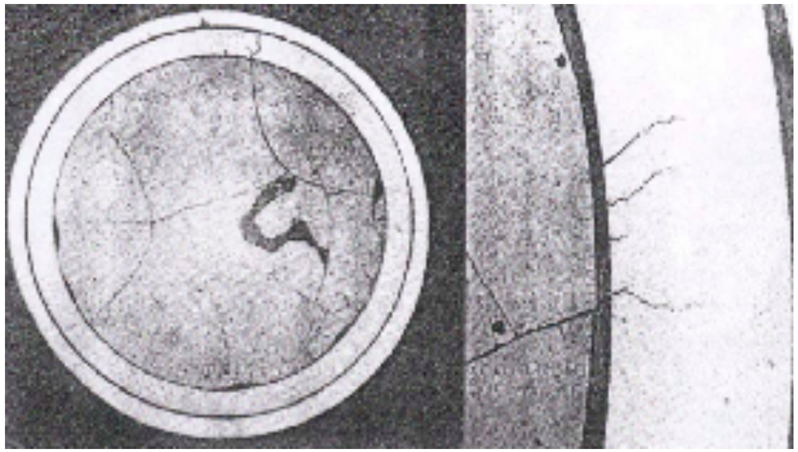
Zirconium alloy research challenges

Corrosion



Delayed hydride cracking

Pellet Cladding Interaction



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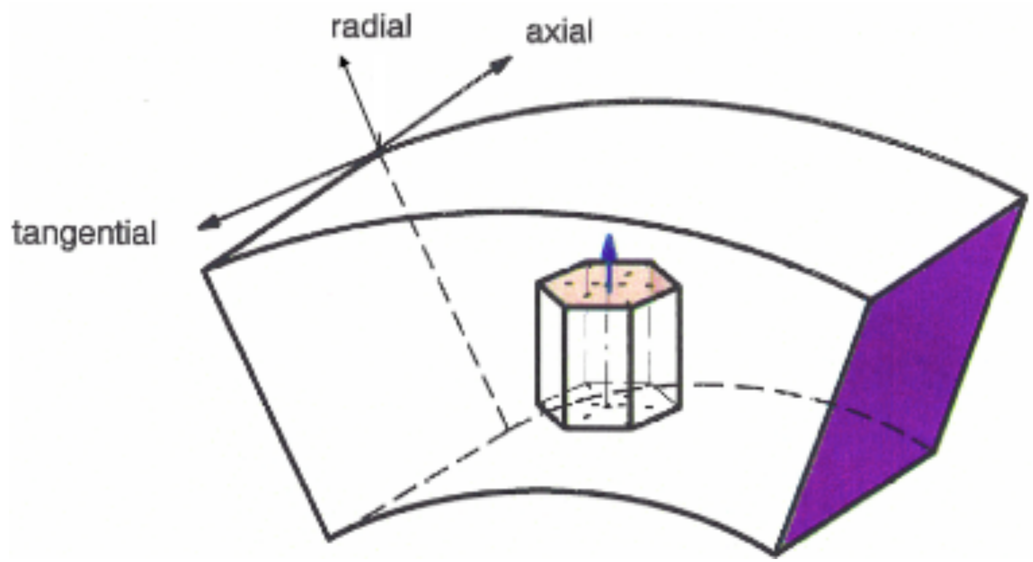
Texture development during pilgering

- Highest stresses are in the tangential direction
- Under deformation, the basal poles align themselves in the direction of the major compressive stresses



(wall thickness) / (diameter) > 1 :

c-axis will preferentially orientate towards the radial direction

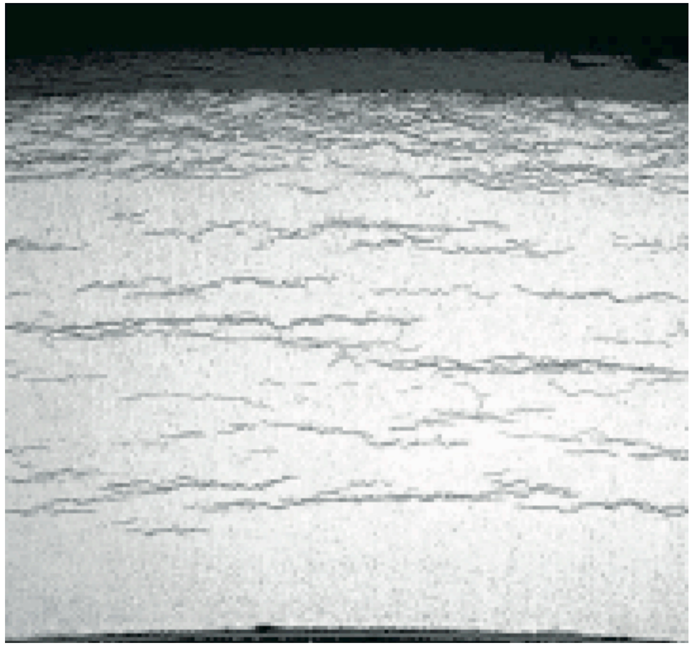
(wall thickness) / (diameter) < 1 :

c-axis will preferentially orientate towards the tangential direction

⇒ Important consequences for hydride formation, PCI and gap formation between pellet and cladding

Hydrides in Zircaloy cladding: effects of microstructure

Recrystallised microstructure



Stress relieved microstructure

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Delayed Hydride Cracking - DHC

- In recent years, cracks have been observed in high burnup LWR fuel rods originating from the outside of the cladding (i.e. not PCI cracks)
- Similar phenomenon observed over many years in PHWR pressure tubes
- Hydrogen diffuses to region of high tensile stresses (i.e in front of the crack tip) an precipitates out
- Hydrides grow and crack due to large stresses
- Crack propagation rate determined by diffusion rate of hydrogen

⇒ **DHC also of importance for dry storage**

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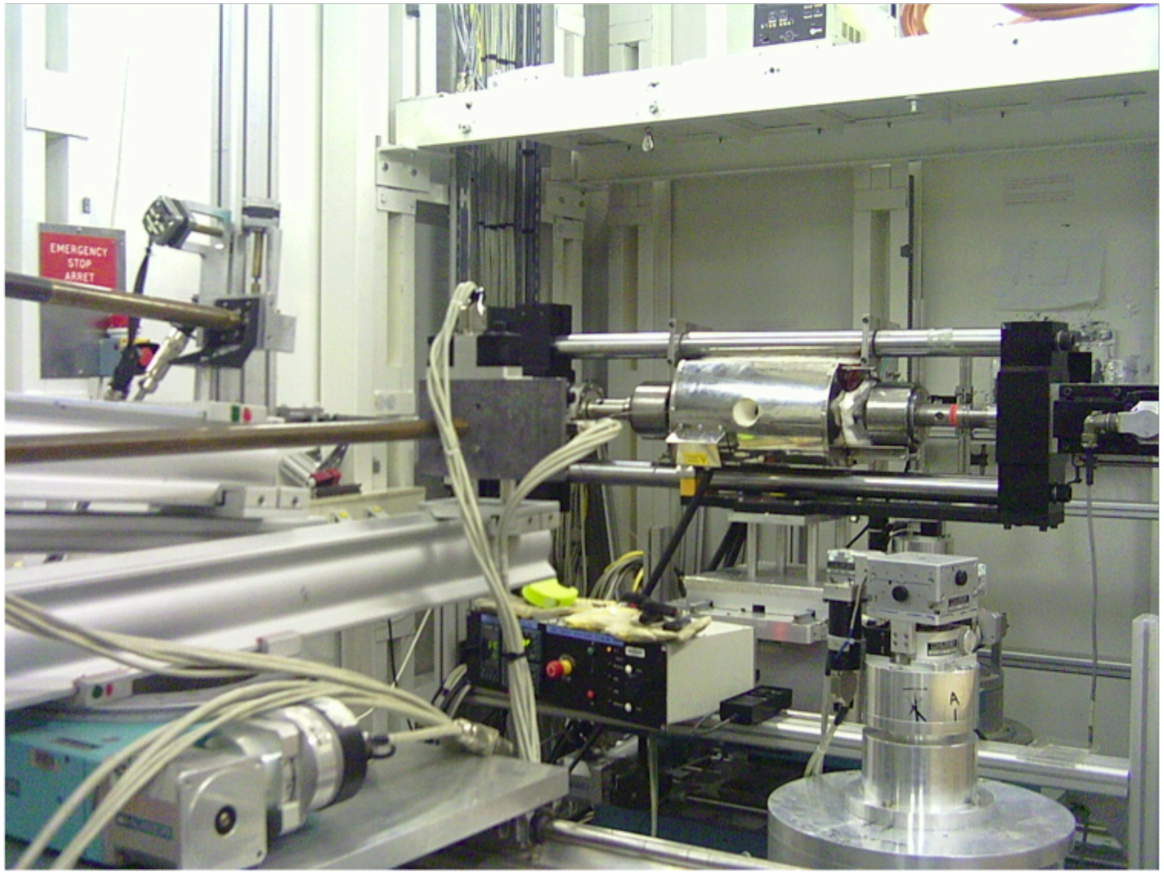
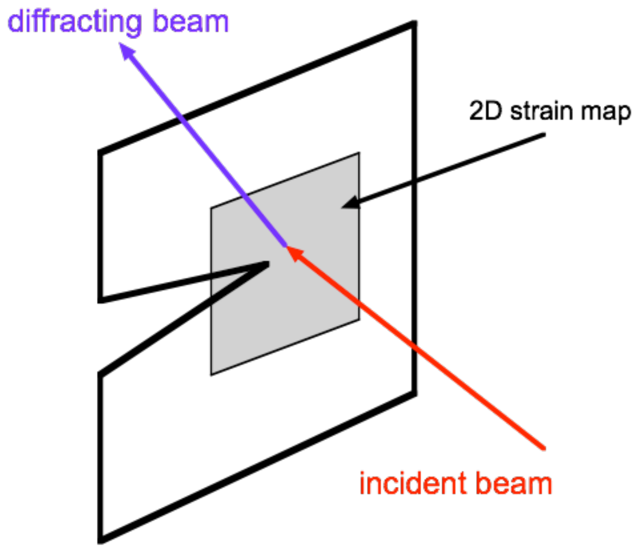
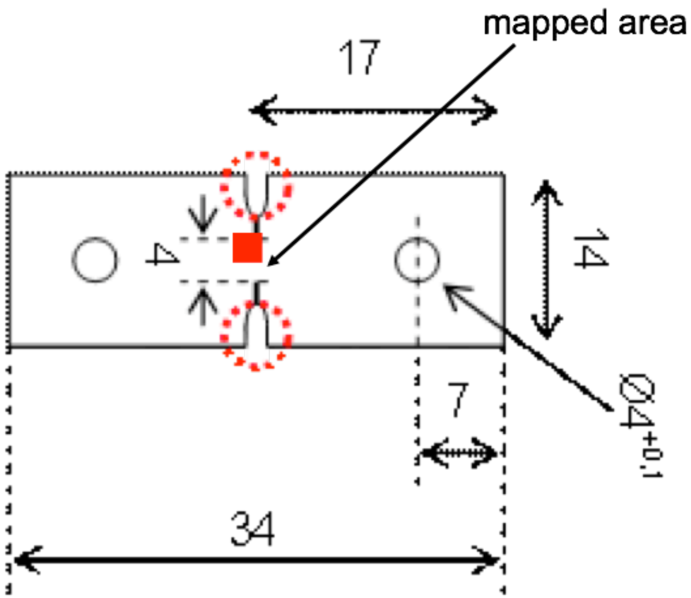
Mapping strain during DHC







Source: M Preuss

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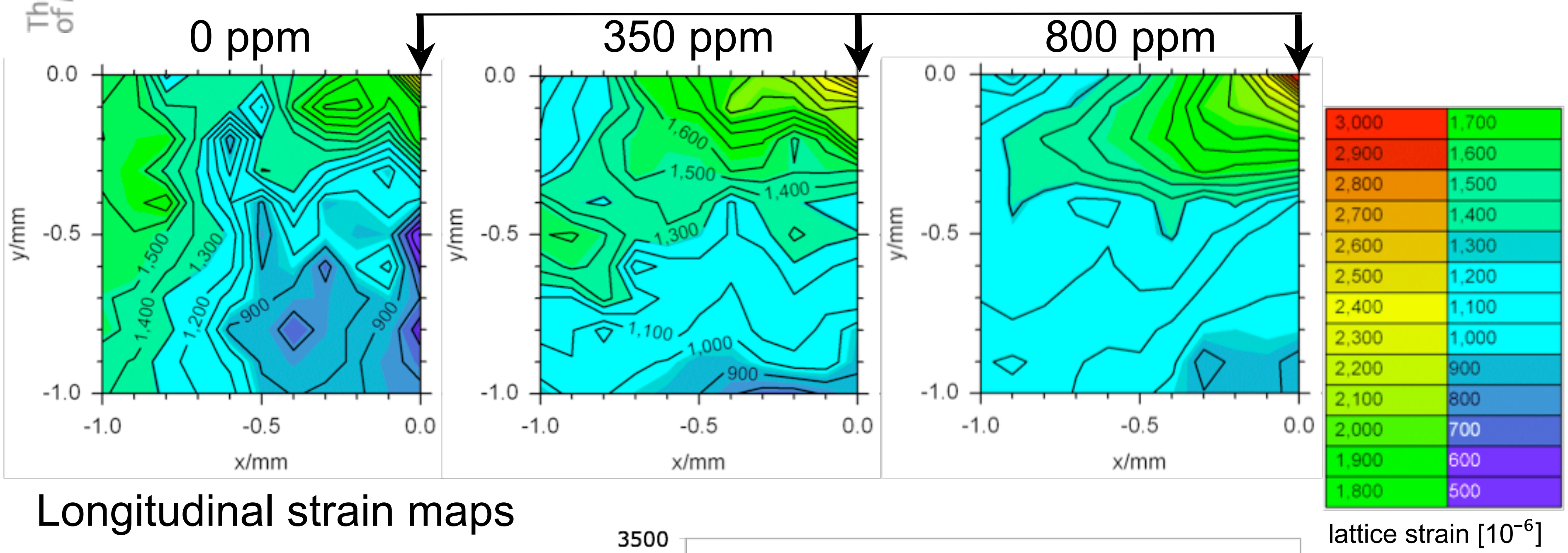
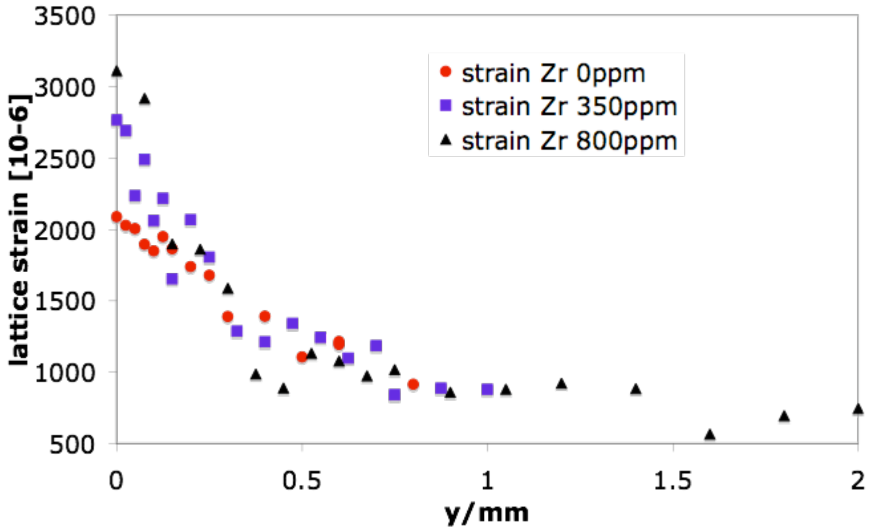
Strain maps (20°C and 150 MPa)



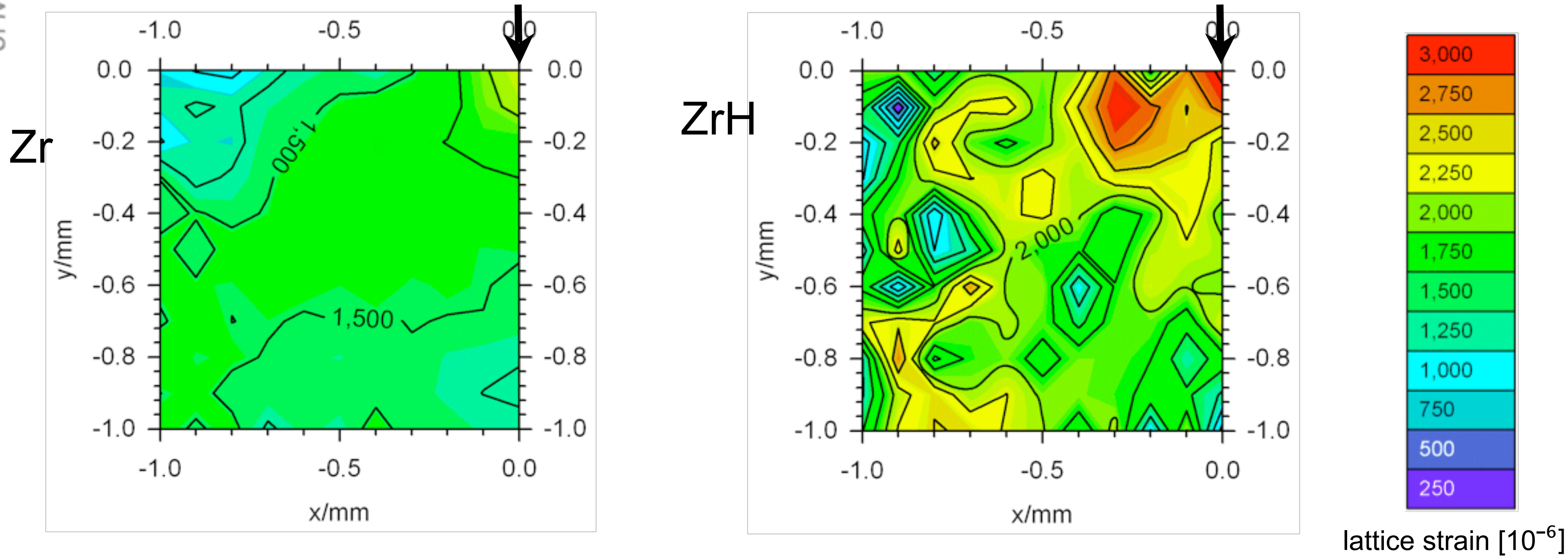
Longitudinal strain maps

Strain profiles
(x = 0 mm)



Source: M Preuss

Strain maps at 400ºC and 150 MPa

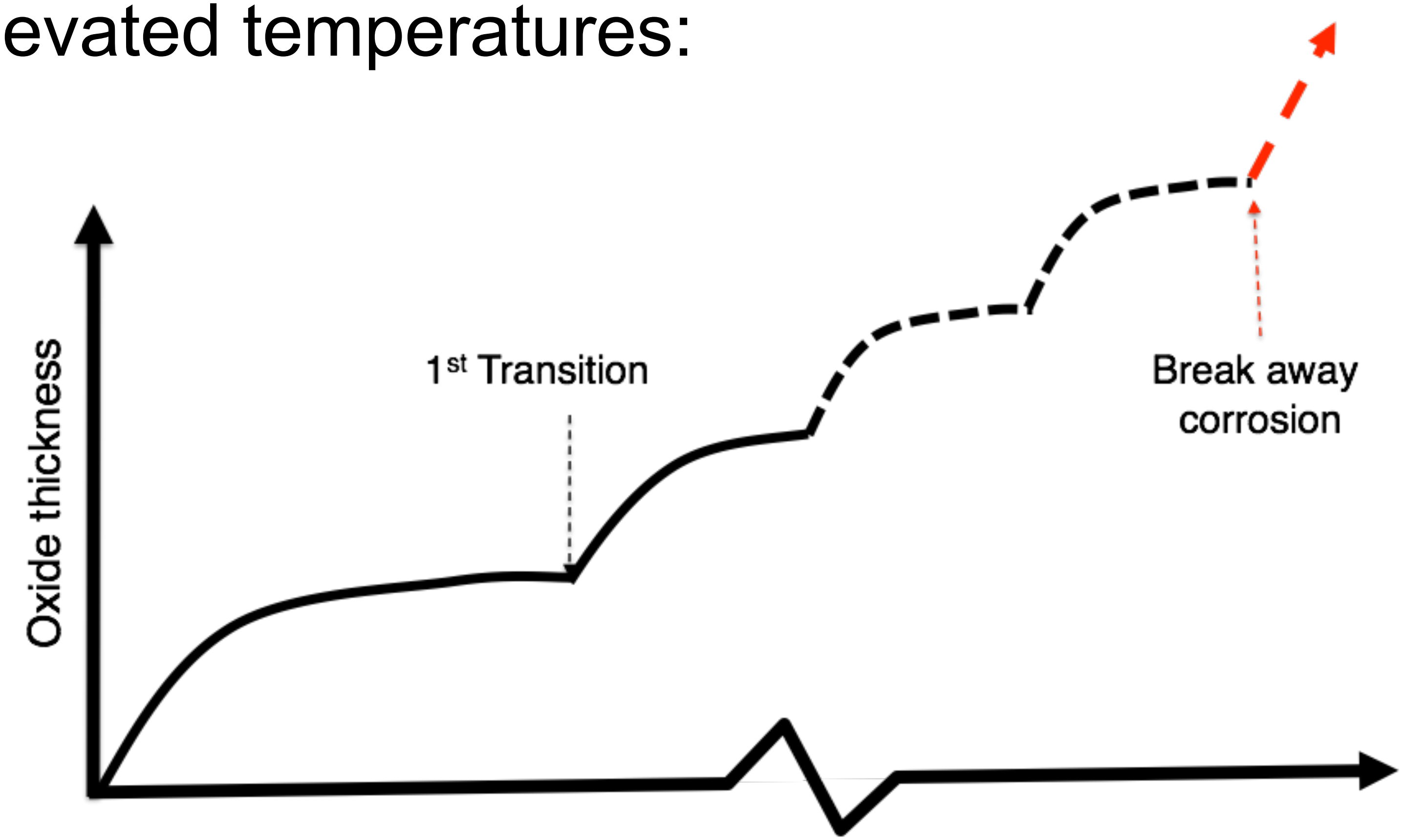


δ-Hydride (cubic) seems to gradually transform to γ-hydride (tetragonal) during deformation, which would be associated with ordering of the hydrogen

Source: M Preuss

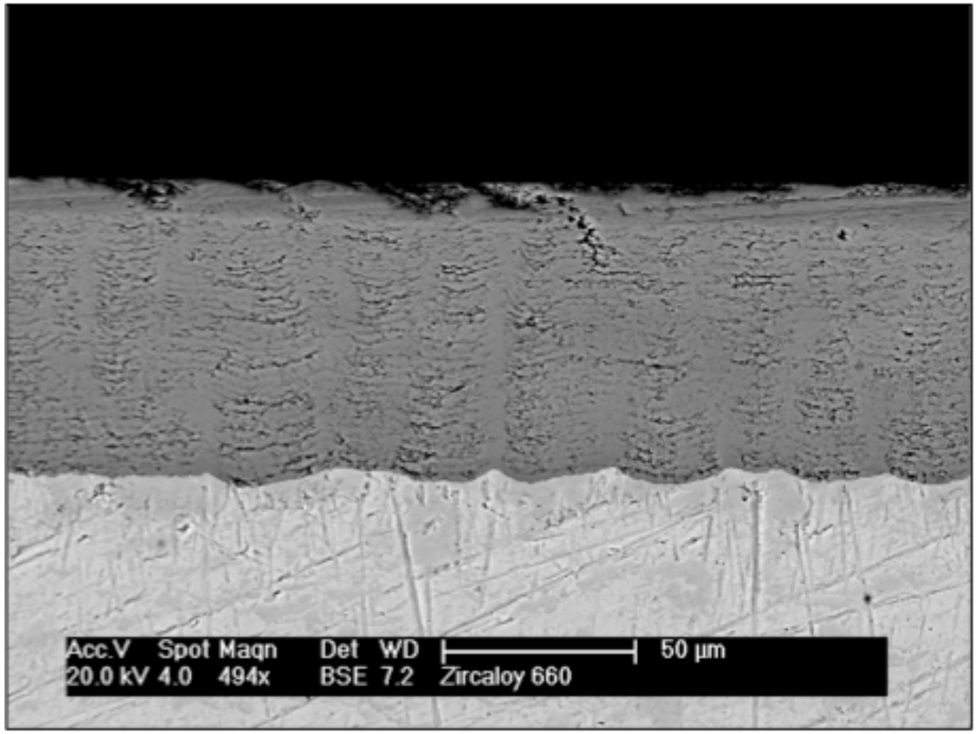
Corrosion mechanisms in Zr-alloys

Pre- and post-transition corrosion in Zr-alloys at elevated temperatures:

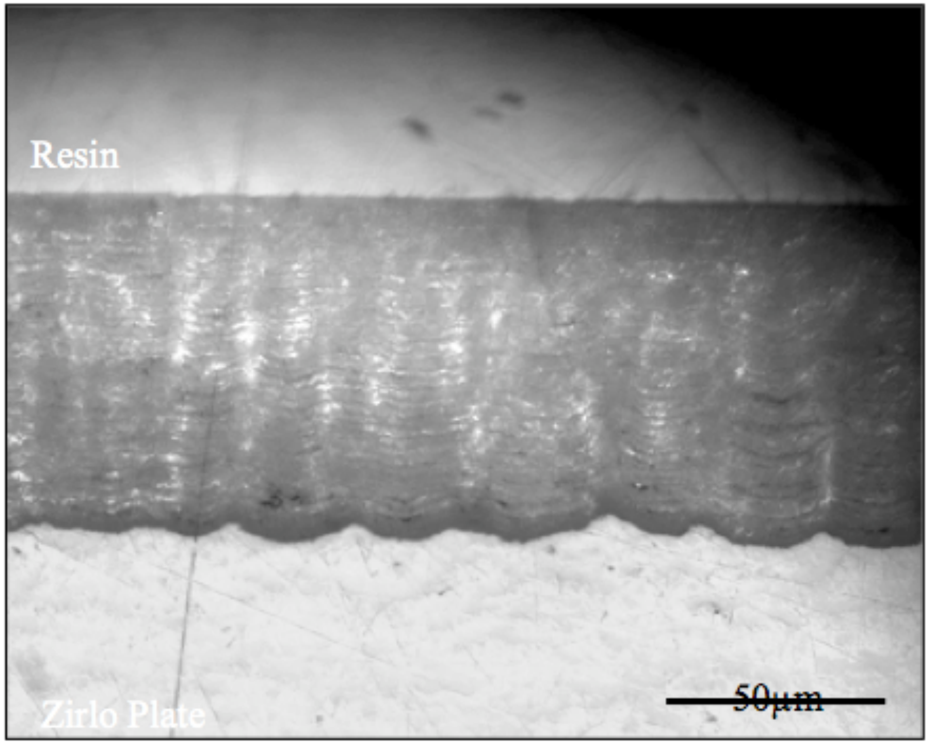


Layered oxide structure

- large compressive stresses and stoichiometric variations in the oxide stabilise tetragonal ZrO_2
- as corrosion proceeds, oxide remote from the interface starts to transform into the stable monoclinic phase
- phase transformation induces cracking, leading to enhanced corrosion



Backscatter SEM image



Transmission optical micrograph

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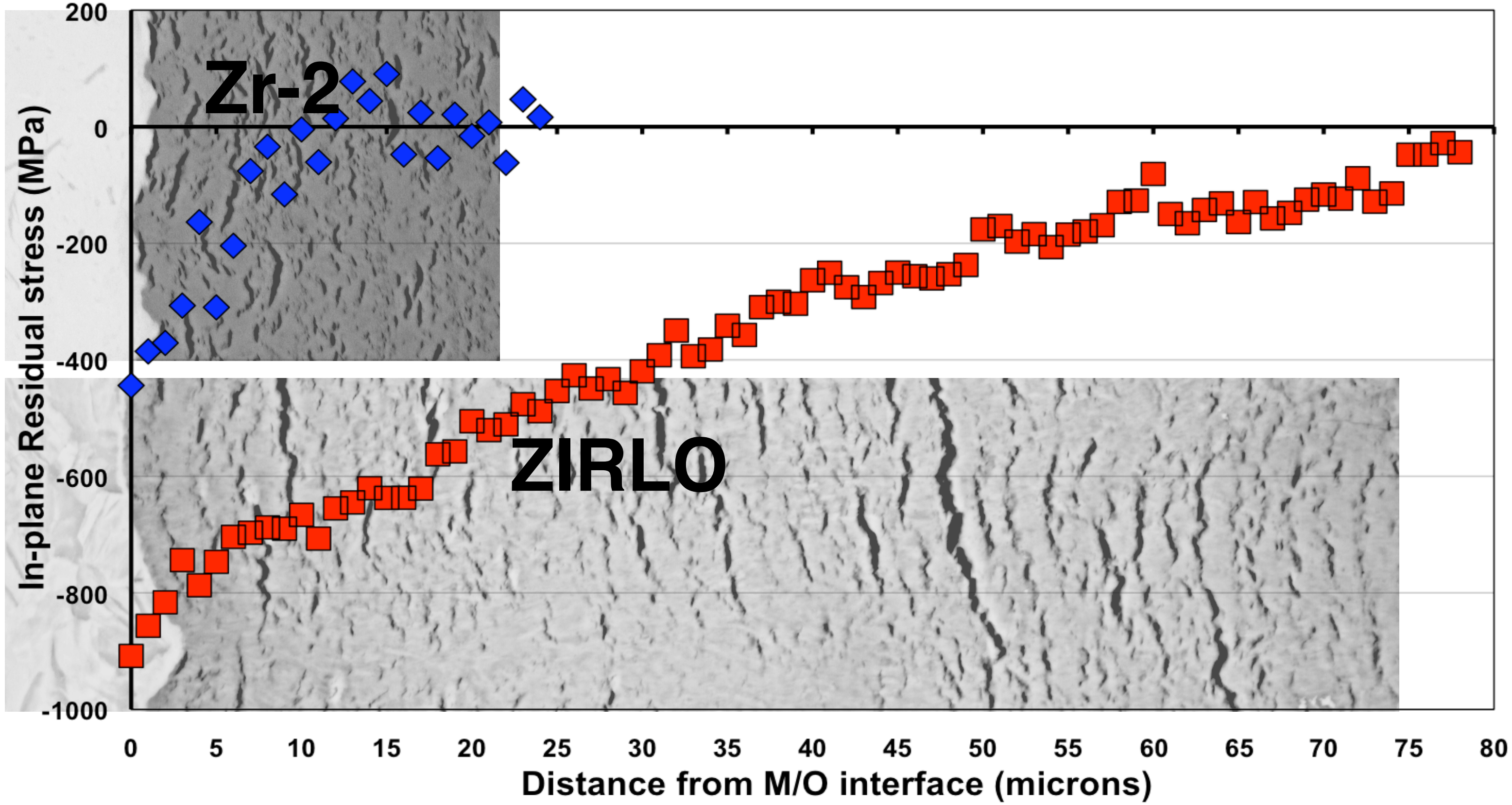
Residuals stresses: Zr-2 vs. ZIRLO



- Do residual stresses in the oxide affect:
  - oxidation kinetics ?
  - tetragonal to monoclinic phase transformation ?
  - initiation of oxide cracking / promote porosity ?

Structural Materials

- Structural Materials should :
  - Have sufficient strength
  - Not deteriorate due to irradiation and/or corrosion
  - Maintain geometry within acceptable limits
  - Not become excessively activated by neutrons (e.g. cobalt in steel giving ^{60}Co)
  - Have a low cross-section for neutron absorptions (less important outside of the active core)

Fuel material - metals

- Metal fuel relatively easy to fabricate
- Good thermal conductivity, moderate melting point
- High density
- Susceptible to radiation induced changes - swelling, distortion, cracking, spalling etc.
- Susceptible to oxidation

Fuel material - ceramic

- Not straightforward to fabricate
- Poor thermal conductivity, but high melting point
- Less susceptible to radiation induced changes - but still an issue
- Lower density than metal
- Fuel cladding should be stable under irradiation, strong, corrosion resistant and have low neutron capture cross-section
  - Common choices Zircaloy, stainless steel

Why is oxide fuel so dominant ?

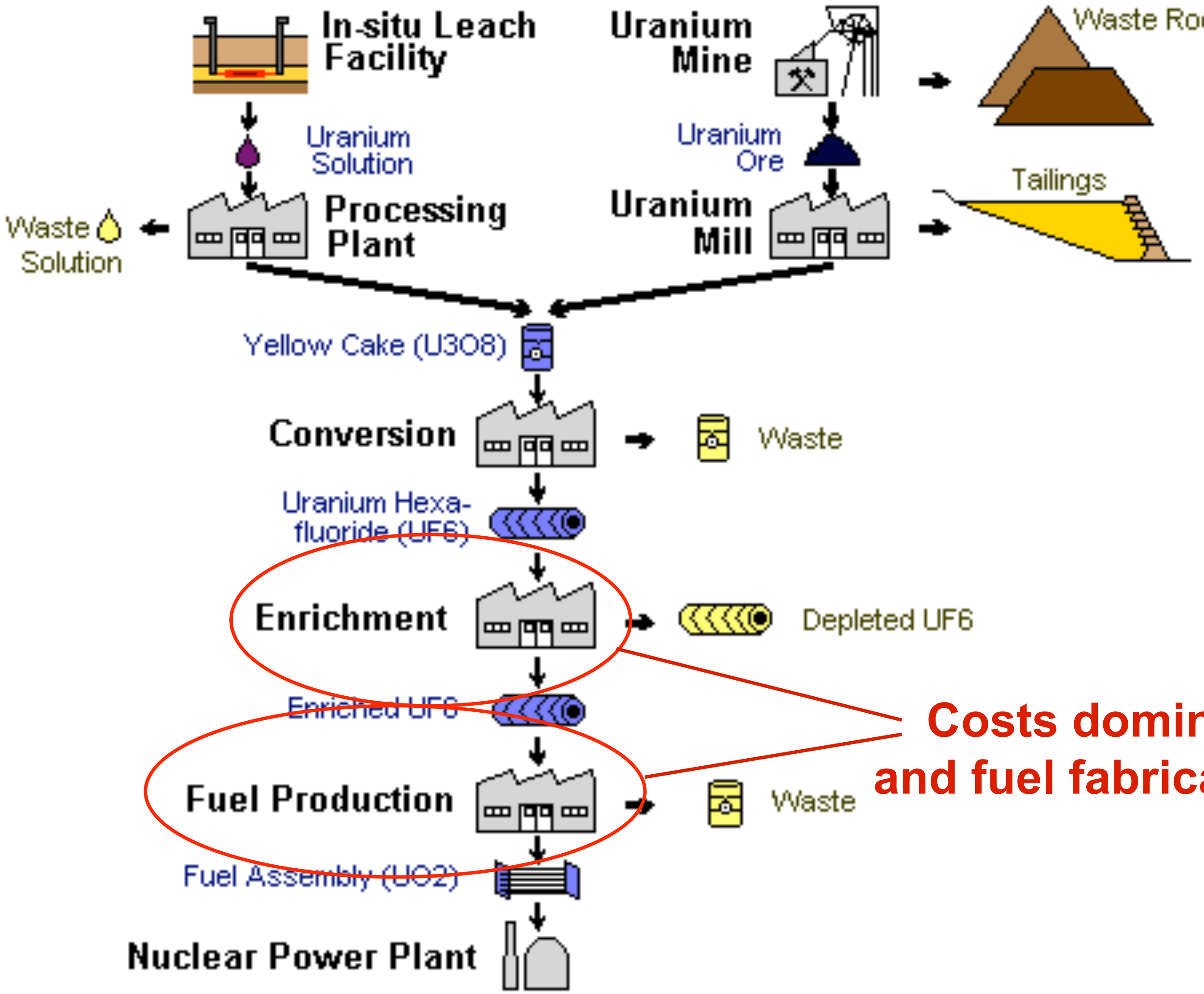
- It has a high melting point (almost 2850°C), compensating for its poor thermal conductivity;
- It is chemically compatible with water, CO_2, sodium (with which it does react, but only slowly), stainless steel, and zirconium alloys.
- It is relatively easy to fabricate into ceramic pellets, and exhibits good physical stability.
- Oxygen has a low neutron capture cross section.
- It is compatible with reprocessing processes, and is stable over long periods (required for long-term storage)
- The huge data base of experience with oxide makes the implementation of alternatives difficult

Metal, carbide, nitride, cermet, cercer and inert matrix fuels

- Metal fuels historically used in Magnox/UNGG and fast reactors
- Carbide fuels tested in fast reactors and of interest for advanced fast reactors
- Nitride fuels of interest for advanced fast reactors
- CerMet involves a mix of ceramic and metal
  - eg UO_2 granules in a molybdenum matrix
- CerCer a mix of two ceramics
- Inert matrix fuels have no fertile isotopes (e.g. ^{238}U)
  - No new fissile materials from fertile captures
  - High rates of Pu and minor actinide destruction, but practical limitations because of poor thermal properties

Fuel manufacturing

Fuel cycle: front end



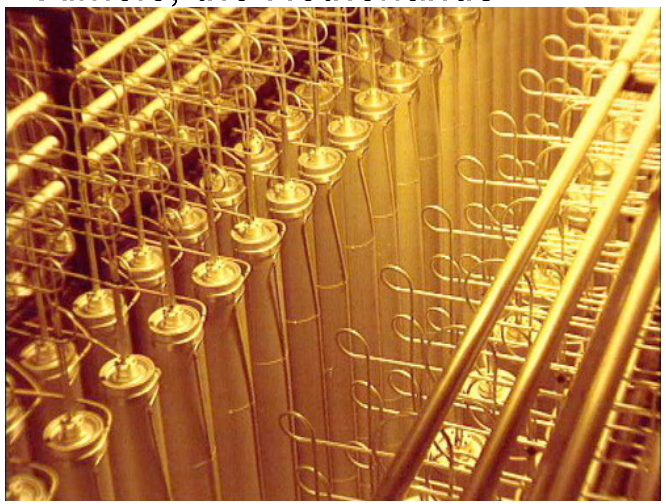
Costs dominated by enrichment and fuel fabrication

Enrichment

- Legacy plants based on gaseous diffusion
  - Very large plant, high capital cost
  - Large process inventory
  - High operating costs (2,500 kW.h / SWU)
  - Large amount of waste heat
  - High operating cost
  - Relatively inflexible
- Modern plants based on gas centrifuges
  - Power consumption < 5% of equivalent diffusion plant (approx. 50 kW.h per SWU)
  - Compact layout: low capital cost
  - Lower inventory: flexible performance
  - Future plants may employ laser separation (SILEX, etc.)

George Besse diffusion plant Tricastin, France

URENCO centrifuge plant Almelo, the Netherlands

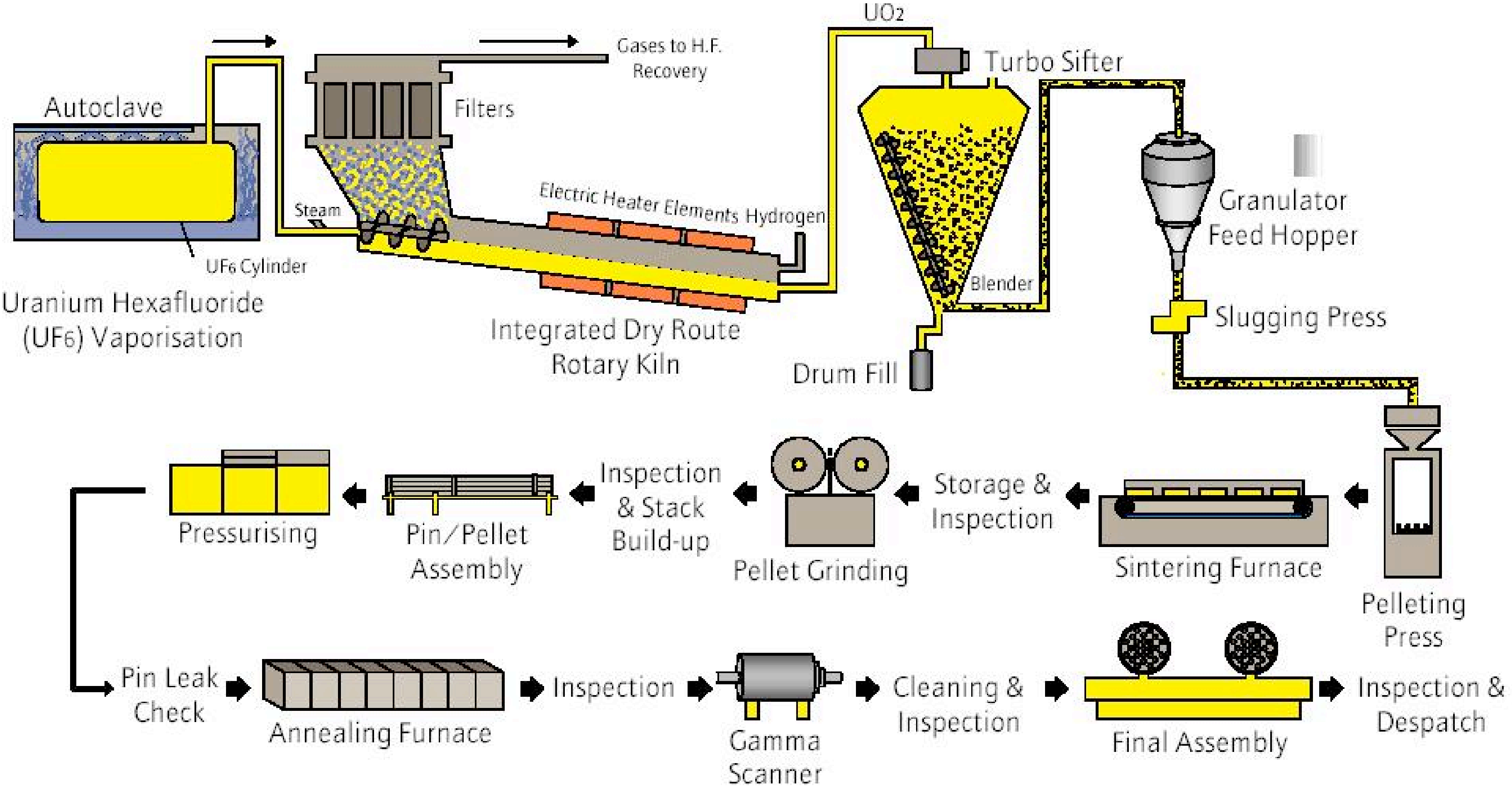


Fuel Manufacture

- Chemical processes:
  - Conversion from UF_6 to UO_2
    - ADU (ammonium diuranate)
      - Hydrolysis of UF_6 with ammonia to form ADU
      - Collection and drying of precipitate
      - Pyrolysis at 800ºC followed by reduction with H_2 to give UO_2
      - Very sensitive to the amount of ammonia used in hydrolysis
    - AUC (ammonium uranyl carbonate)
      - Precipitation of AUC by aqueous combination of UF_6, NH_3 and CO_2 gases
      - Reduction of AUC to UO_2
    - IDR (integrated dry route)
  - Scrap recovery (usually by AUC or ADU)

- Mechanical processes:
  - Pelleting
  - Rod build
  - Component manufacture
  - Assembly build

Fuel manufacture by IDR



- IDR provides a simpler processing route than ADU or AUC, with essentially no liquid effluents
- Original BNFL process has been further optimised, e.g. by Areva

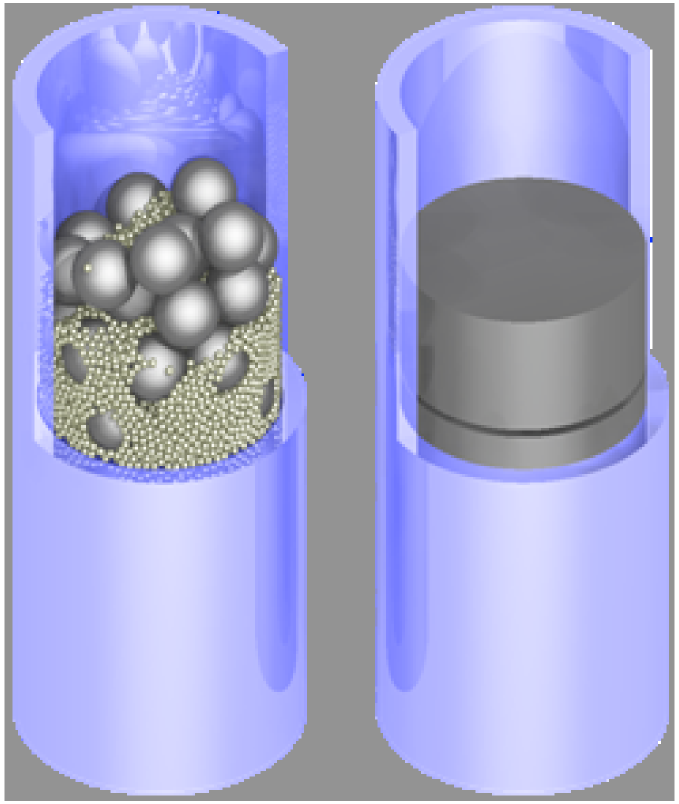
Source: BNFL

Alternative fabrication methods

- Vibro-pack
  - Avoids high-precision pelleting and grinding by using vibration to compact granules into the rod
  - Fewer residues
  - Suitable for remote fabrication
  - But low packing density
  - Effective thermal conductivity is low
  - Used to produce fast reactor fuel (Russia)

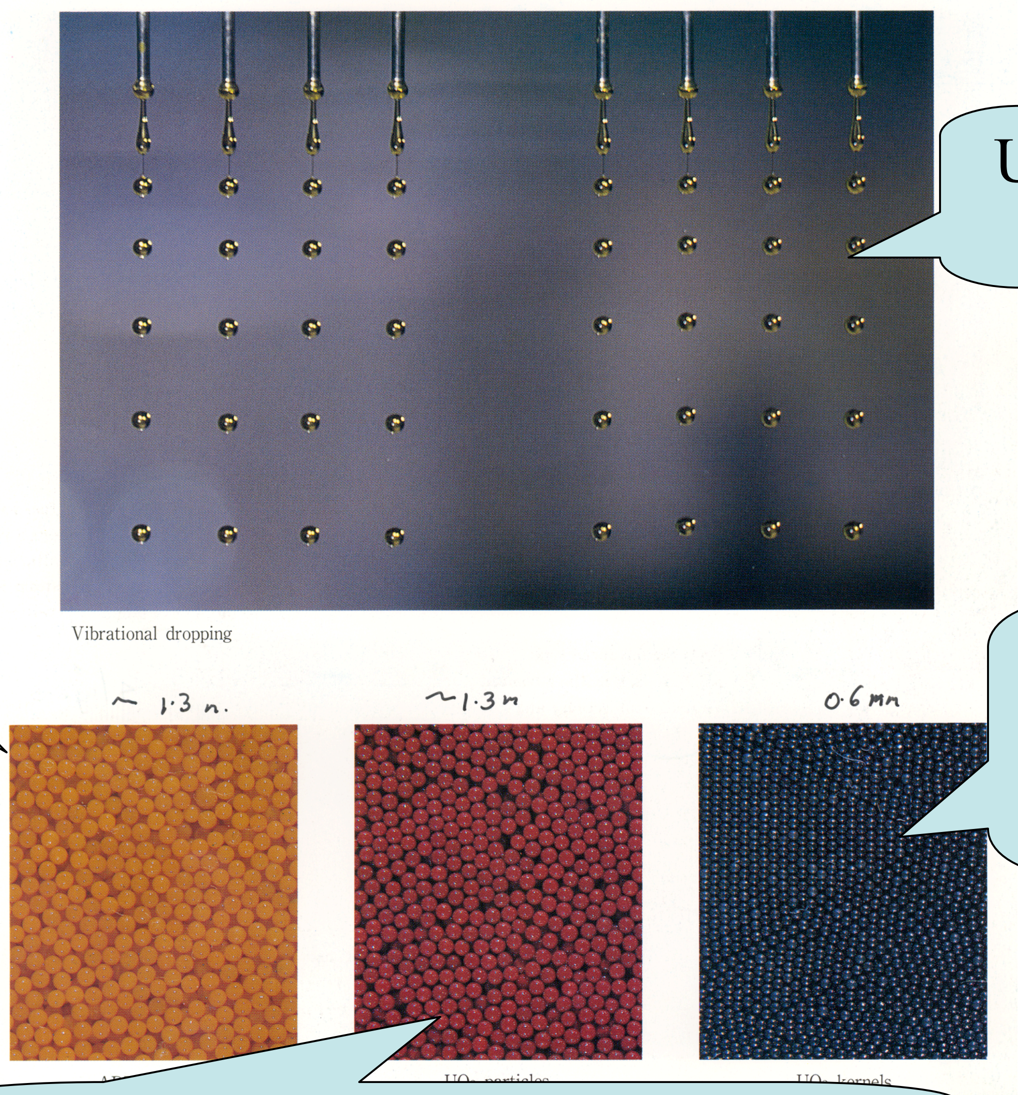
Alternative fabrication methods

- Sol gel then sphere-pac
  - Wet process produces very uniform fuel spheres
  - 2 or 3 sphere sizes to get good packing fraction
  - Spheres vibro-packed
  - Dust free
- disadvantages
  - produces liquid effluents
  - low effective conductivity
  - low density
  - Used to produce fast reactor fuel (UK)



Source: Paul Scherrer Institute

Sphere-pac kernel manufacture



Uranyl Nitrate droplets

ADU particles

Sintering to produce UO_2 kernels

Calcined to produce UO_3 particles

In-reactor performance:

What happens to fuel in a reactor?

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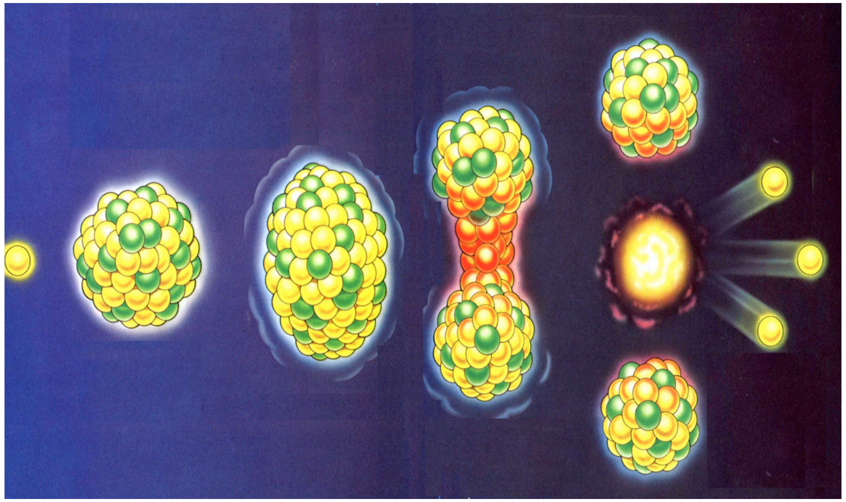
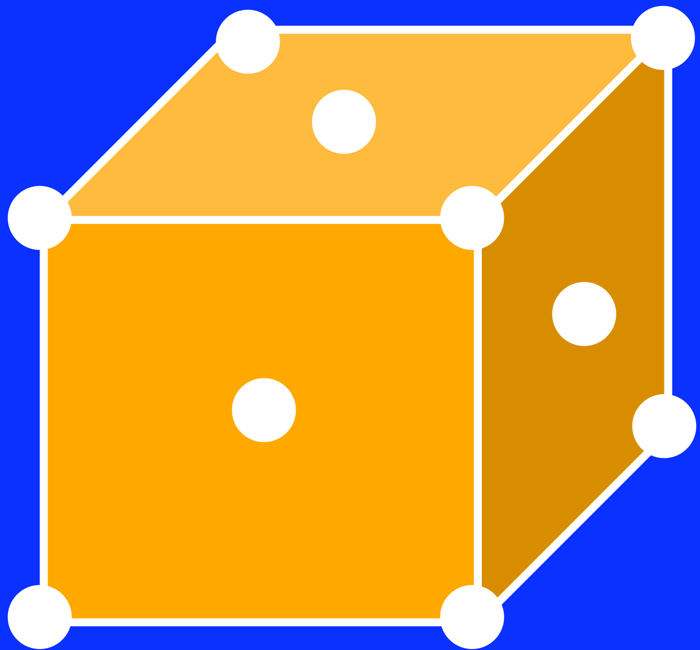
It undergoes fission and produces energy

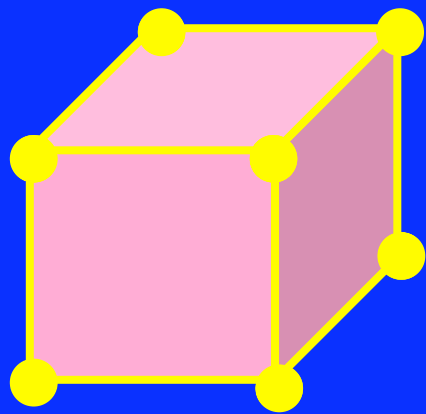


- Each fission produces 2 (sometimes 3) fission fragments, plus 2 or 3 fast neutrons
- Fragments known as a ”fission spike”
  - 6 μm in length, 10 nanometres diameter
  - 10 pico-second duration
- Every atom affected within 3 mins, and disrupted ~ 500,000 times during life
- Power production:
  - 10 fissions per cubic micron per second
  - one pellet ~ 6 trillion ($6x10^{12}$) fissions/second
  - total energy released per fission is about **200 MeV** (32×10^{-12} J)
    - kinetic energy of fission fragments 82%
    - kinetic energy of free neutrons 2%
    - gamma rays 3%
    - delayed radioactivity 7%
    - neutrinos 6%

UO_2 fluorite crystal structure



- U^{4+} anions
- face centred cubic
- lattice ~ 0.56 nm

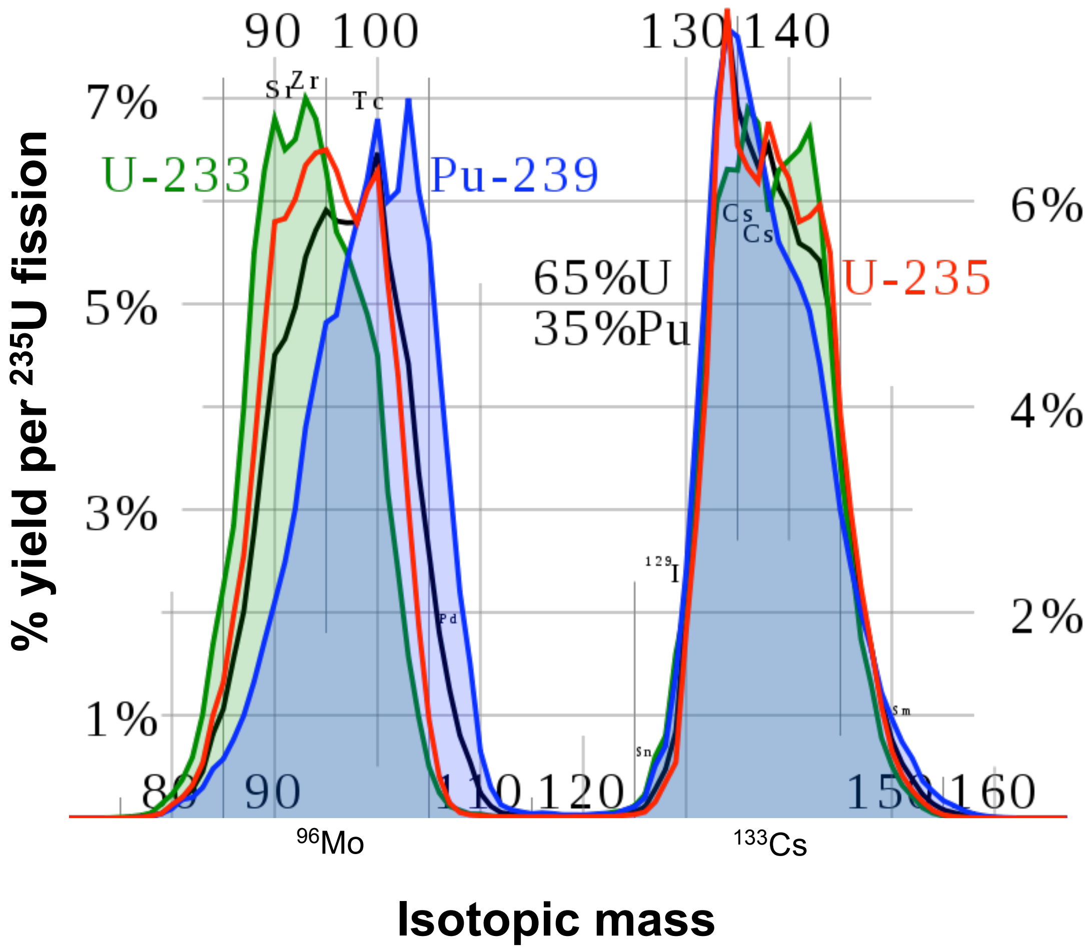


- O^{2-} anions
- simple cubic
- lattice ~ 0.28 nm

It undergoes fission

- Fission fragments fly apart in opposite directions
  - highly charged ions interact very strongly
  - ~200,000 Frenkel pairs on cation (U_4+) lattice
  - ~10,000 Frenkel pairs on anion (O_2-) lattice
- Referred to as a ”fission spike”
  - 6 microns in length, 10 nanometres diameter
  - 10 pico-second duration
- Majority of fuel structural damage results from fission spikes (not neutrons)

Yields from ^{235}U thermal fission



- Y/rare earths 53%
- Zr/Nb 30%
- Ru/Tc/Rh/Pd 26%
- **Xe/Kr 25%**
- Mo 24%
- Cs/Rb 23%
- Ba/Sr 15%
- I/Te 1%
- others 3%
- total ~200%

Source: Forschungzentrum Jülich

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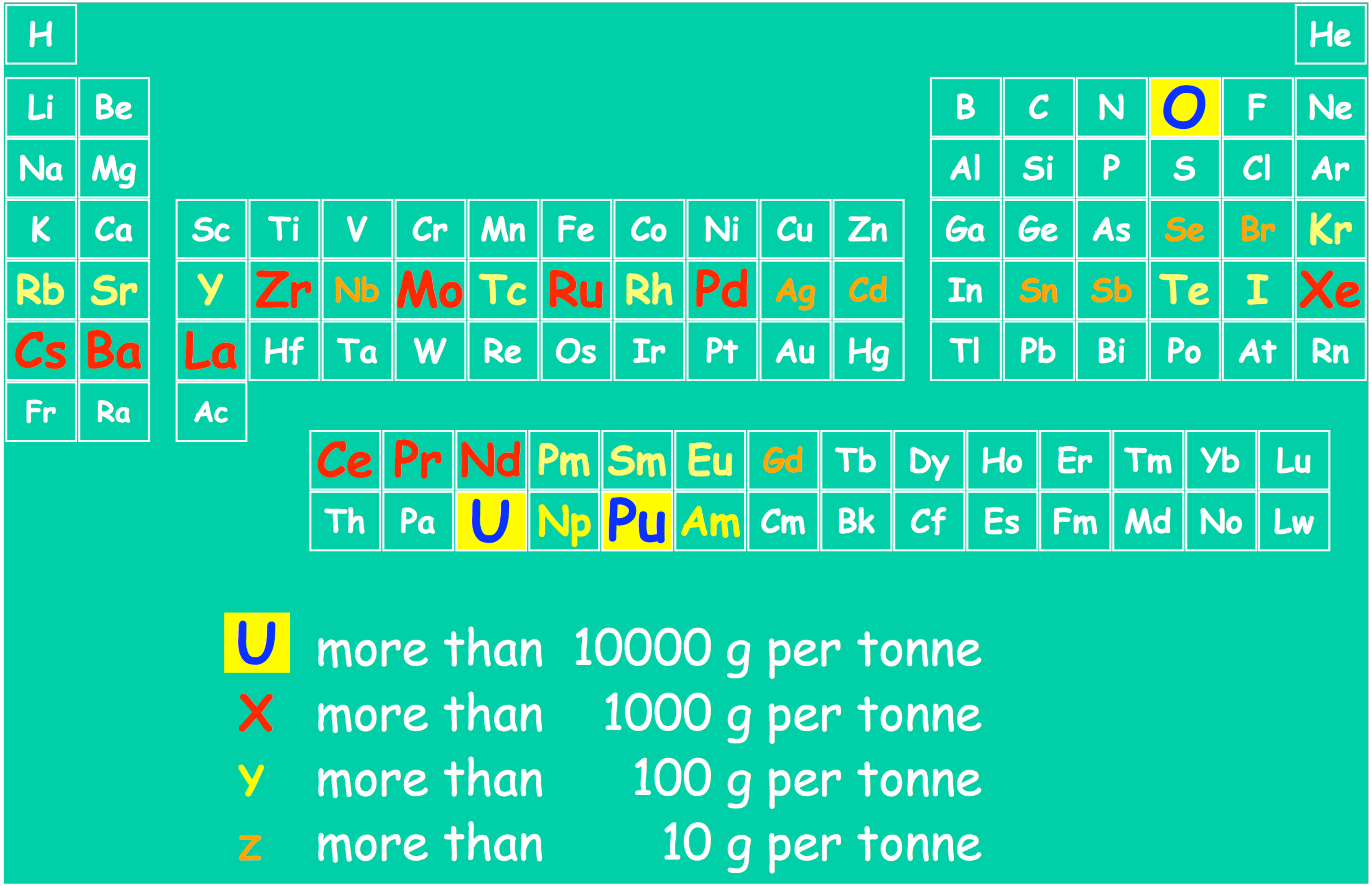
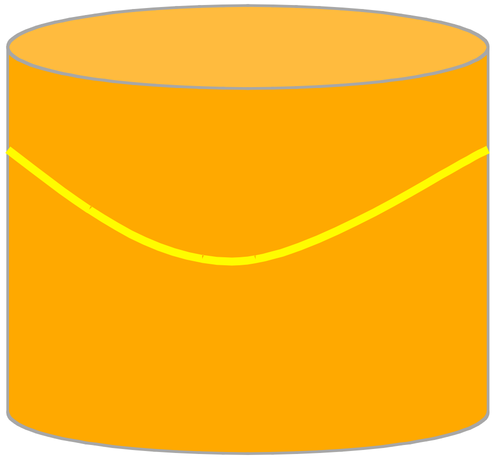
UO_2 composition after 3y in a PWR

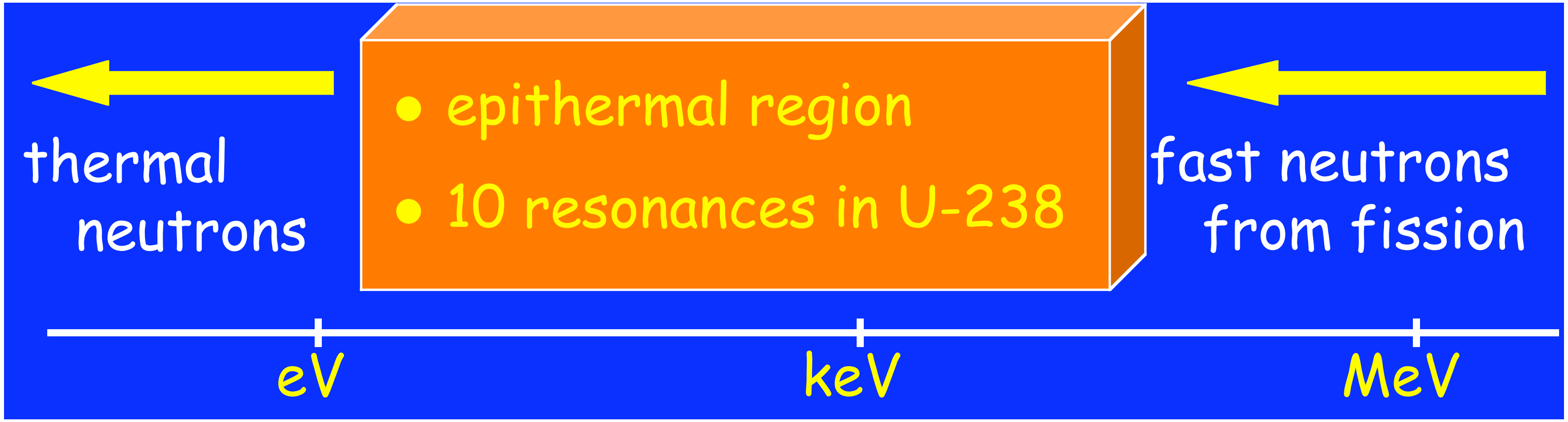


It produces neutrons

- 2 or 3 fast (~ 1 or 2 MeV) neutrons per fission
  - clad corrosion, growth, embrittlement, etc
  - pressure vessel embrittlement, etc
- Neutrons are slowed down in the moderator
  - source of thermal neutrons is outside the fuel
  - implies thermal flux depression across the pellet



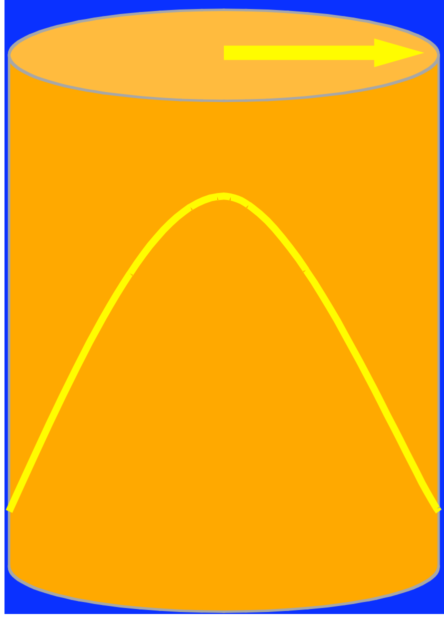
It produces neutrons



- 10 to 20 collisions from fast to thermal energies
  - high capture probability in U-238 resonances
  - U-238 -> U-239 -> Np-239 -> Pu-239
  - plutonium production peaks at pellet surface
  - power depression increases as burnup proceeds leads to “rim effect” at high burnups

It gets hot

- Long, circular cylinder, with ~ uniform power generation
  - negligible circumferential and axial temperature gradients
  - all heat flows radially outwards (1D problem)
  - temperature highest at pellet centre



typical values for a PWR rod

bulk coolant	330 °C
ΔT across coolant film	30 °C
ΔT across clad	20 °C
ΔT across fuel-clad gap	120 °C
fuel outer temperature	500 °C
ΔT across pellet	500 °C
fuel centre temperature	1000 °C

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Estimating Pellet Temperature Rise

simple rule-of-thumb

$$\int_{\text{surface}}^{\text{centre}} k \cdot dT = \frac{Q}{4 \cdot \pi}$$

$Q = 20$ kW/m

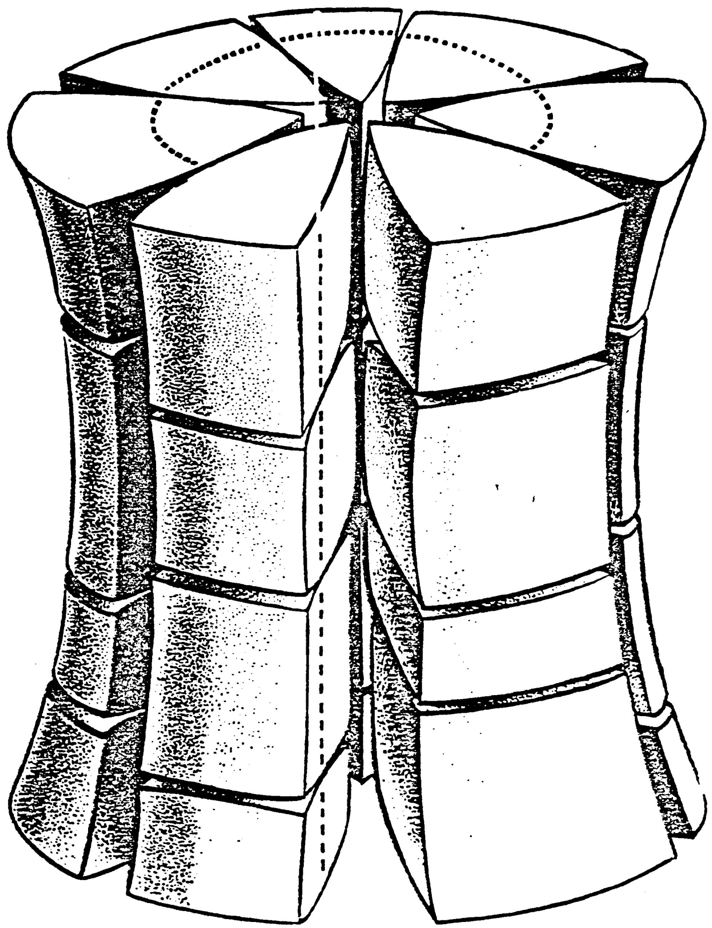
$k = 3$ W/m/K

$$\Delta T = \frac{Q}{4 \cdot \pi \cdot k}$$

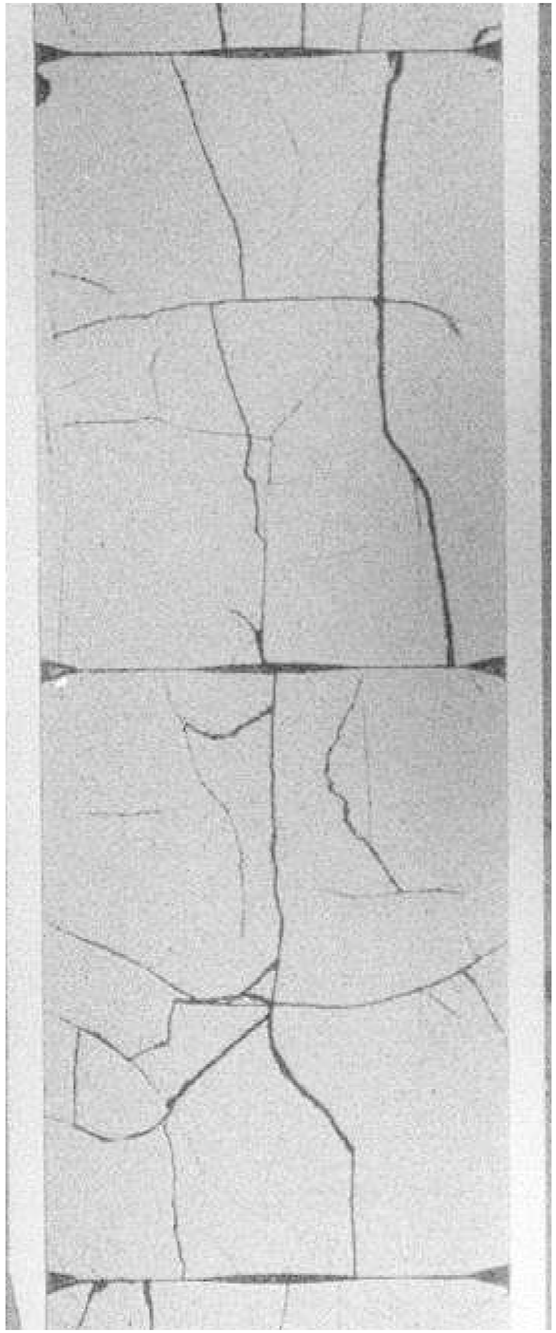
$\Delta T \sim 500\ ^\circ C$

It expands and cracks

- Pellet expands by thermal expansion
- Temperature gradient imposes tensile stresses, i.e. centre wants to expand more than the surface
- UO_2 is brittle, so the pellet cracks
- Cracking is in the r-z and r-θ planes
- Leads to characteristic "hourglass" shape

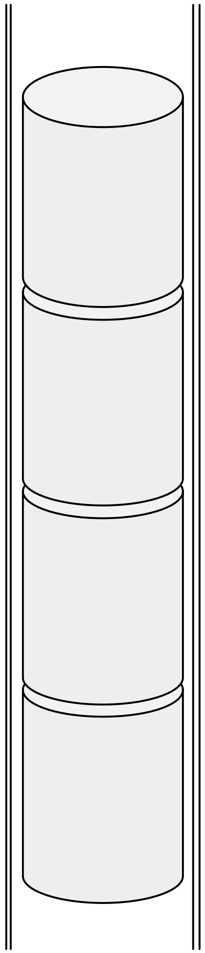


Pellet cracking

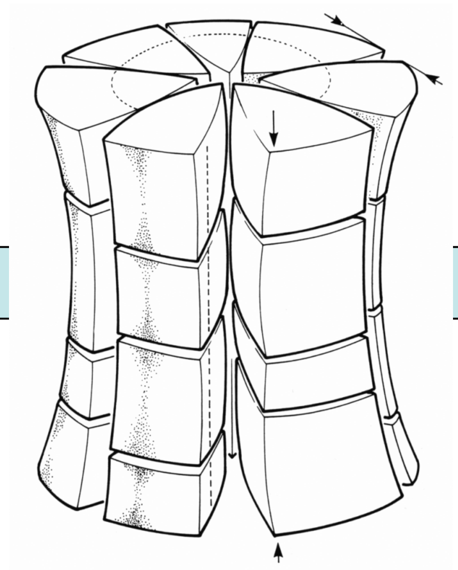


Fuel Response to Irradiation

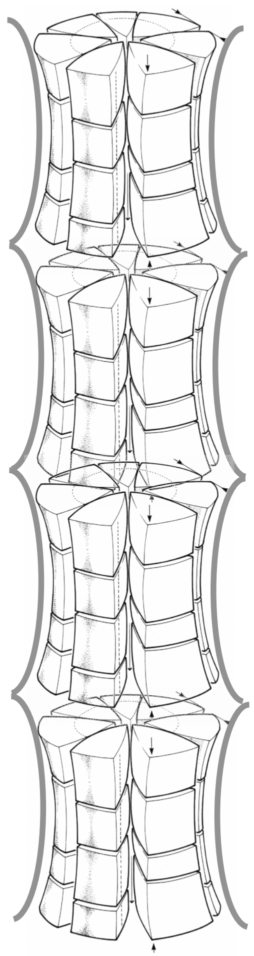
Beginning of life



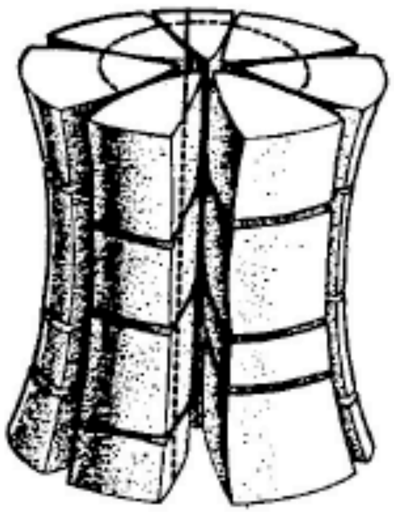
Cracking due to thermal expansion coefficient differences at varying temperatures



After 1 cycle



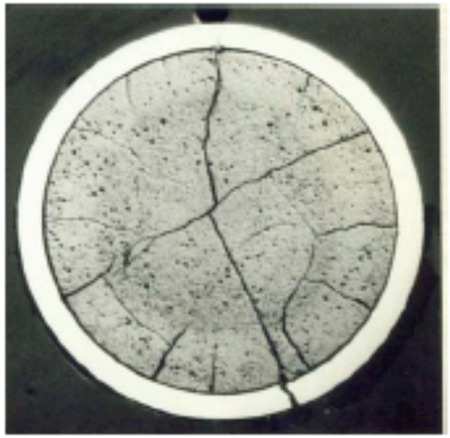
Pellet - Clad Interaction (PCI)

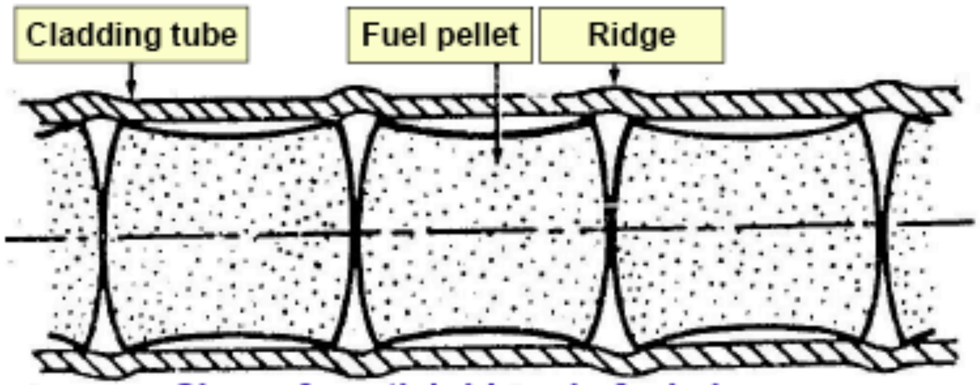

Hour-glassing of fuel pellet due to radial thermal gradient

Longitudinal and radial crack structure develops early in life

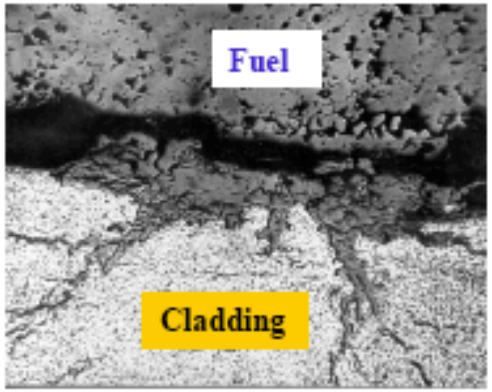
Aggressive fission products accumulate with burnup

Pellet-clad mechanical interaction + corrodant → SCC


PCI/SCC Failure



Circumferential ridges in fuel pins

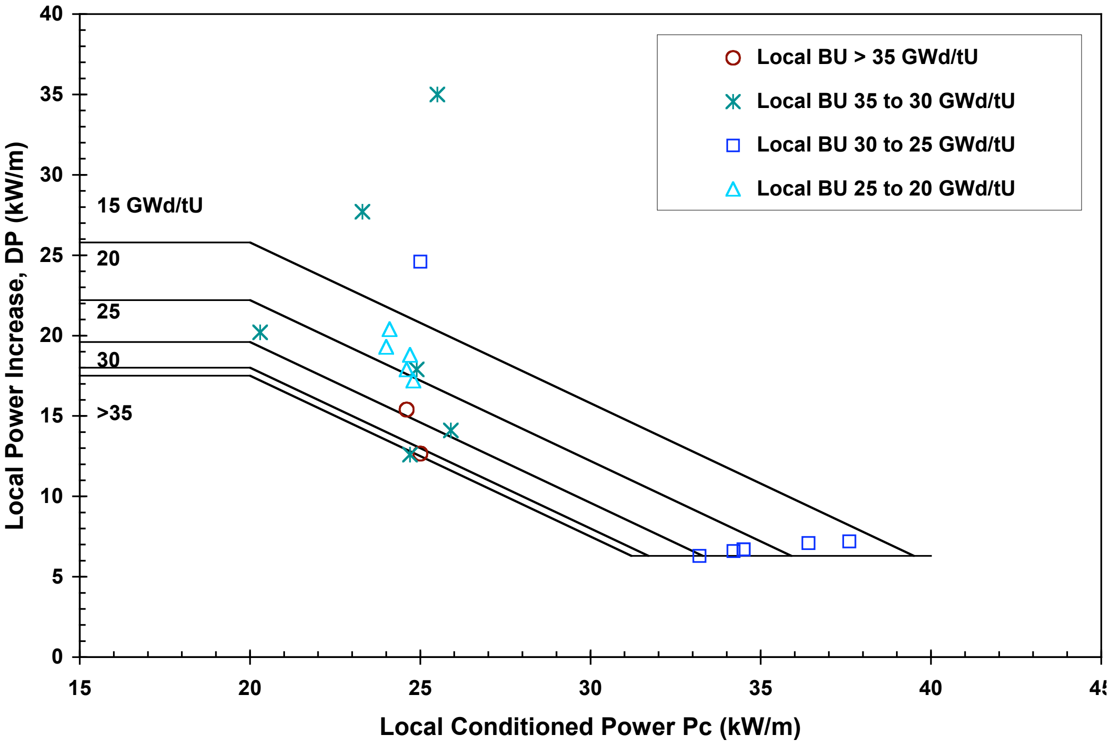


Incipient PCI/SCC cracks

Avoiding PCI failure

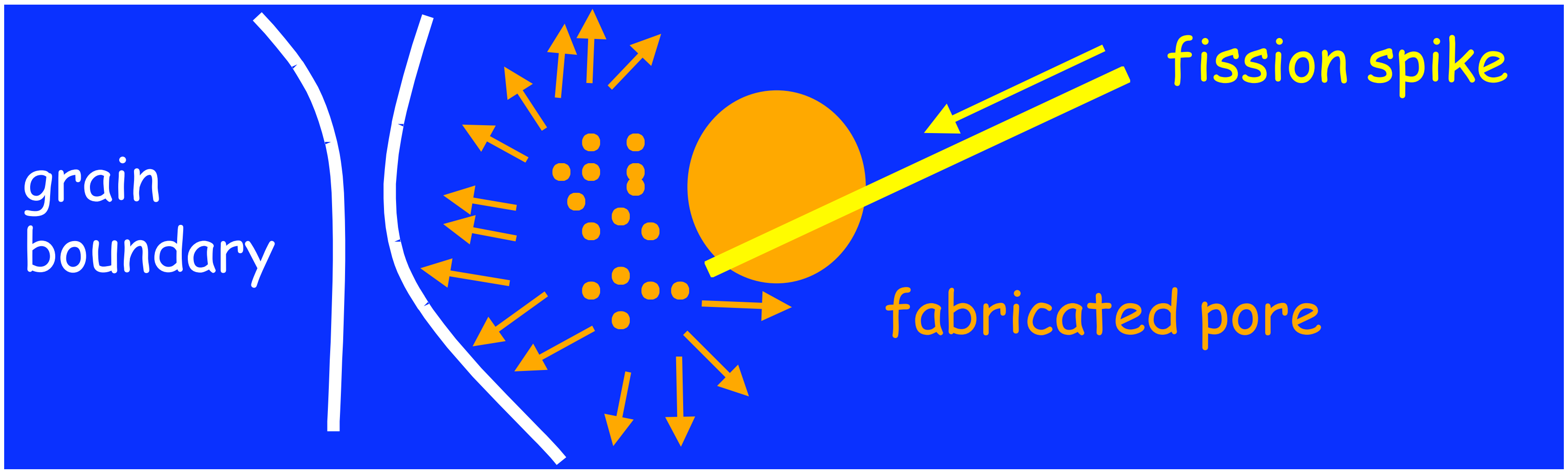
- Many European regulators require an explicit consideration of PCI as a failure mechanism
- Data from ramp tests shows that stress relaxation ("conditioning") influences survival
- Primary creep (thermal and irradiation-induced) plays a major role
- Availability of fission product iodine is a necessary condition

- Data from Studsvik ramp testing (INTER-RAMP, TRANS-RAMP, OVER-RAMP, etc.) has been analysed to determine the cladding stress state prior to and during the power ramps
- Statistical analysis of results yielded a semi-empirical failure model based on "conditioned power", power ramp conditions, and burnup (availability of iodine)



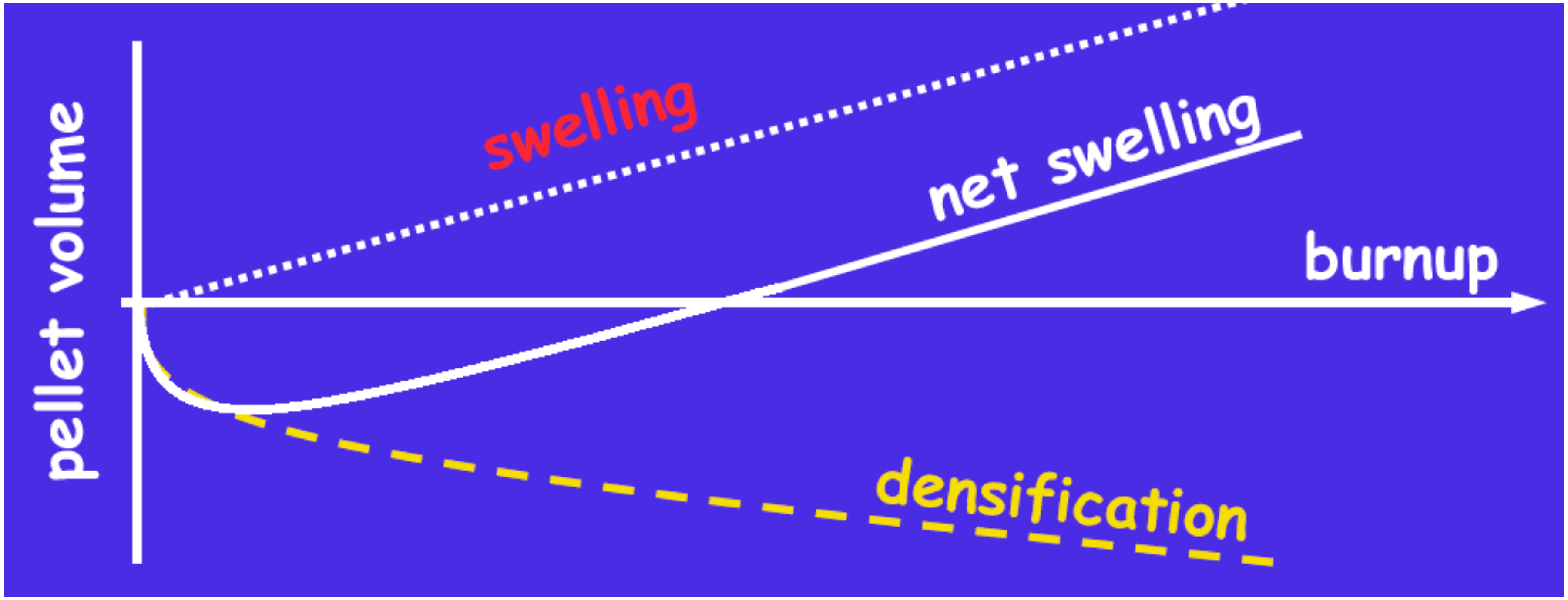
It sinters …

- Small pores (under one micron) are not stable
  - readily destroyed by passing fission fragments
  - typically ~ 1 vol% densification after ~ few weeks
- Larger (spherical) pores are more stable, but are still chipped away by irradiation
  - depends on temperature and grain size



It sinters … then swells

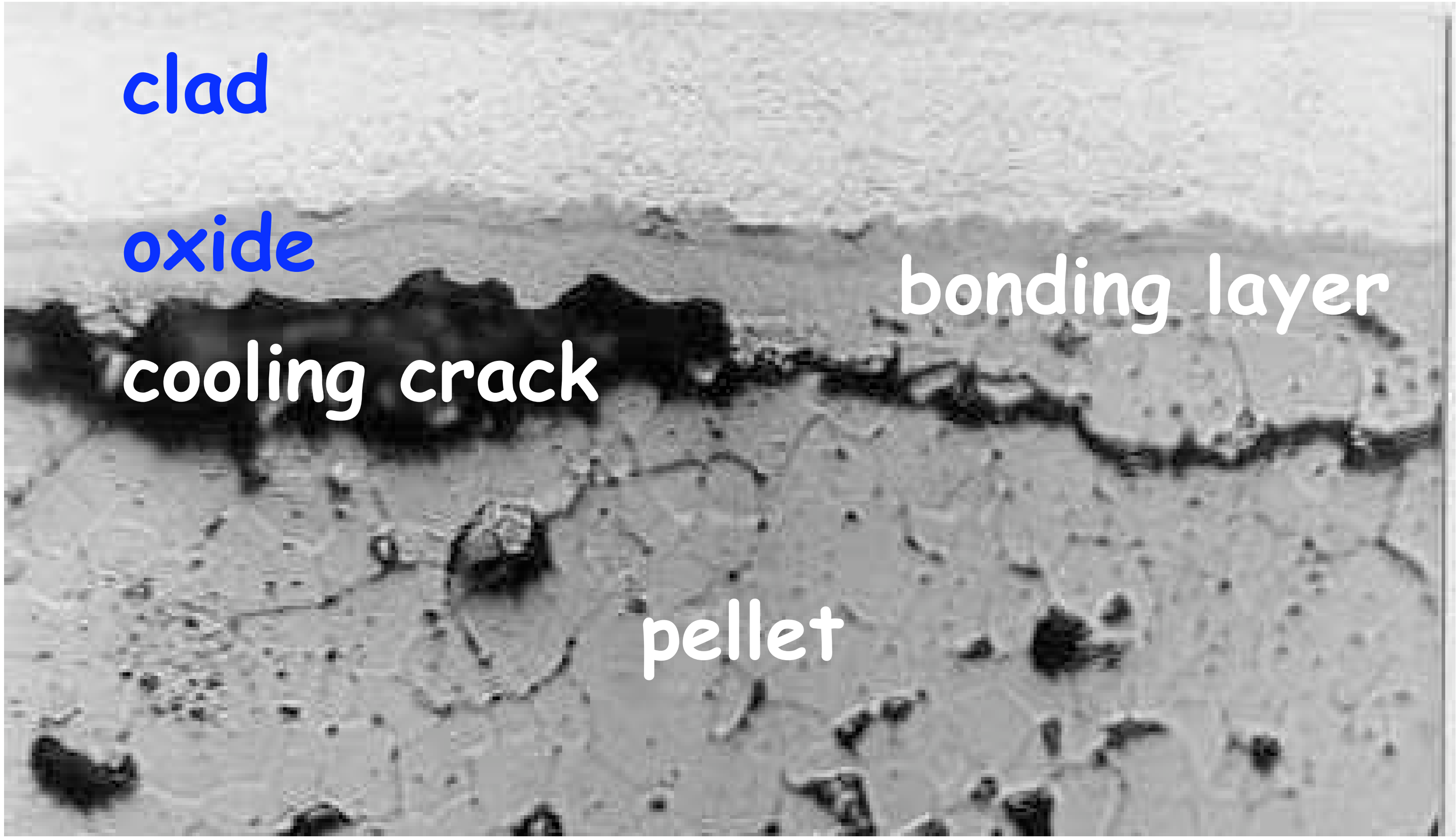
- Many metallic fission products form solid precipitates
- Others are volatile (Cs, I), but solid at low temperature
- Two fission products created for every U that fissions
- Result: volume of fuel increases
  about ½ vol% for every 10 GW•d/tU burnup



It contacts the cladding

- Fuel swelling plus (mainly) clad irradiation creep close the fuel-clad gap after ~1 year in PWR fuel (after a few day in UK AGR fuel)
- Fuel and clad interact chemically and mechanically
  - cladding inner surface oxidises
  - complex interlayer (Zr/U/fp compounds) forms
  - can give fuel-clad bonding at high burnup
  - more significant in MOX which has more free oxygen

… fuel-clad interlayer …



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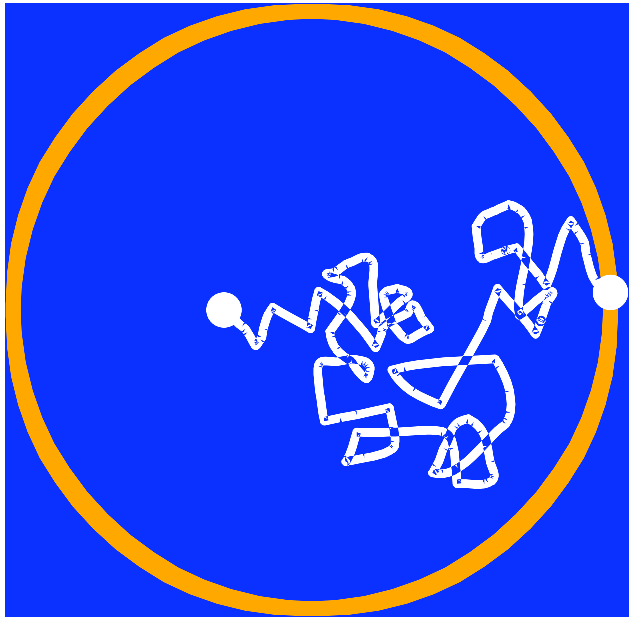
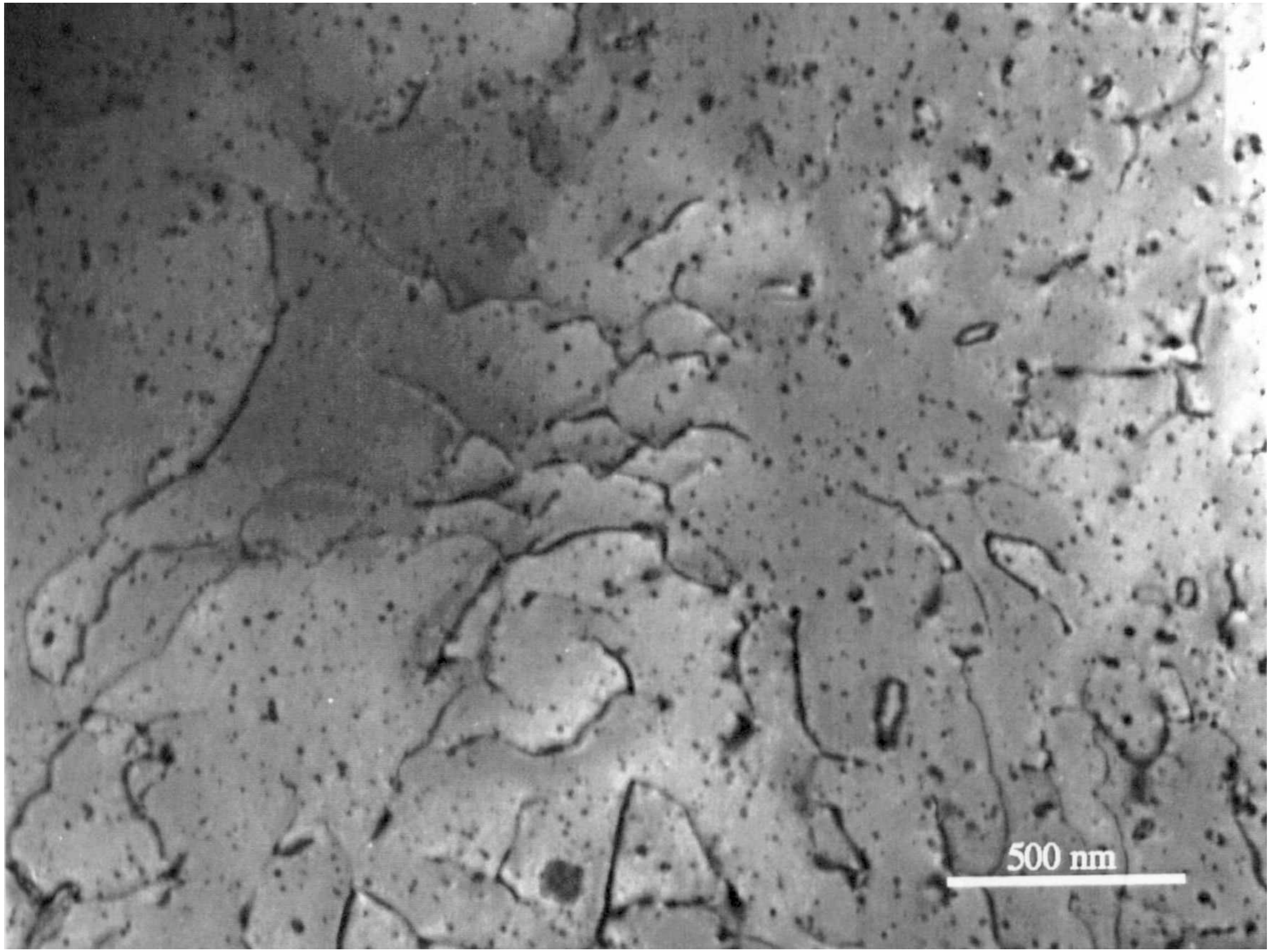
It releases fission gas

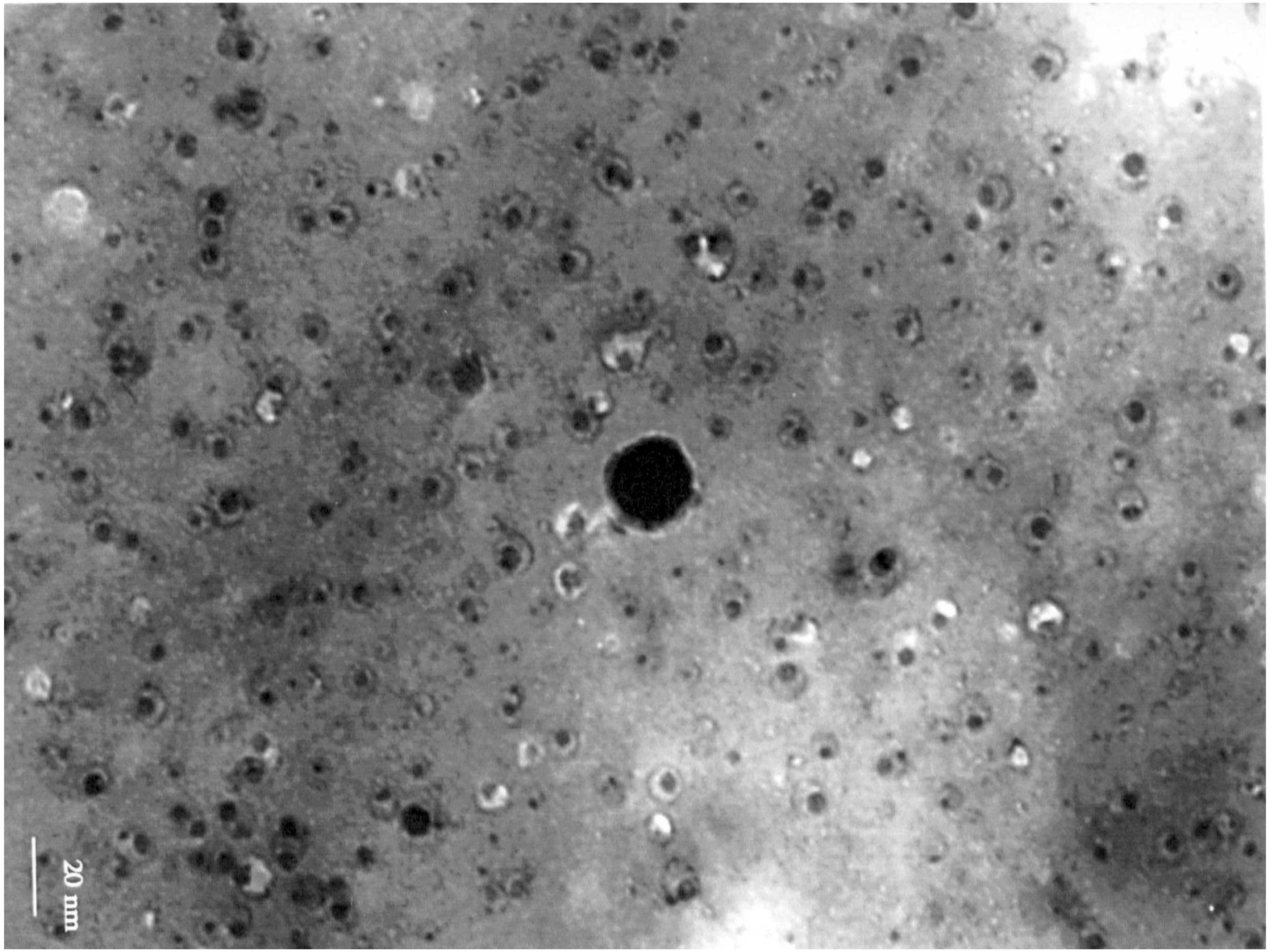
- Xe and Kr have large fission yields
- Insoluble in UO_2 and don't react
- Jump around lattice in 'random walk'
- Depends on thermal energy, i.e. very strong function of temperature
- Diffusion within fuel grains is also affected by
  - inter-granular gas bubbles (which are constantly destroyed and re-nucleated by fission spikes)
  - vacancy lines and loops
  - pinning at inter-metallic precipitates
- Gas may eventually reach grain boundaries



Vacancy lines and loops within a grain

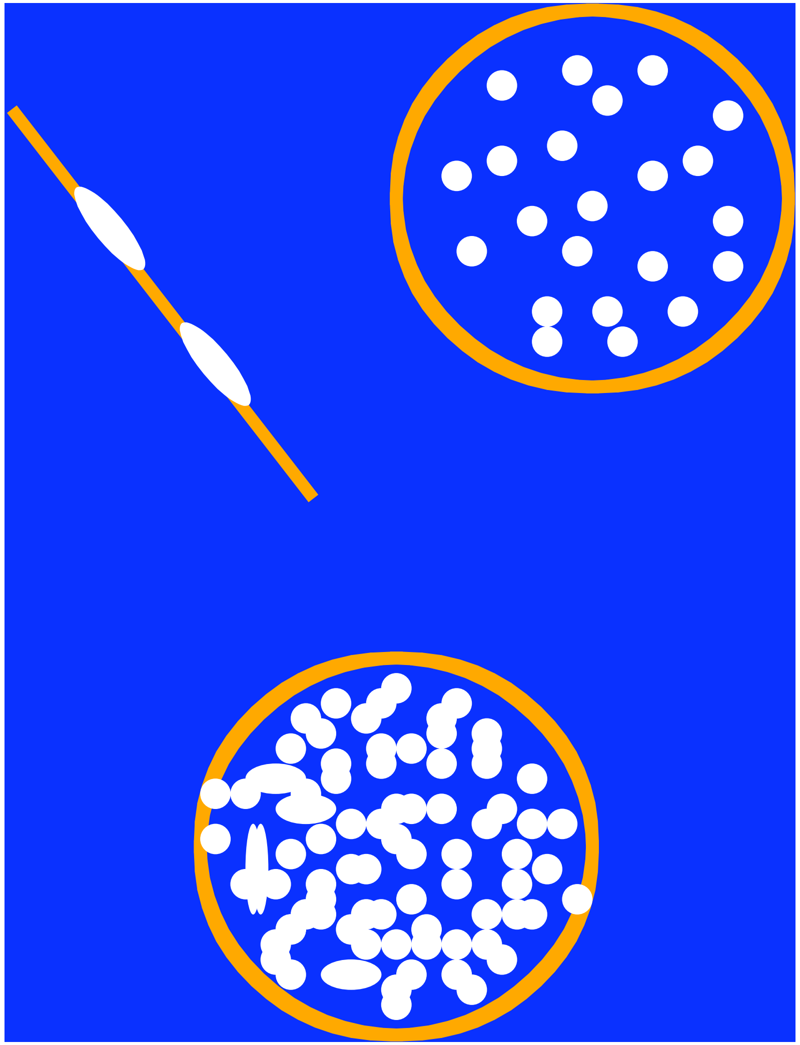


Intra-granular bubbles at high magnification



It releases fission gas

- Lenticular (M&M shaped) bubbles form on the grain faces
- Grain "faces" are where 2 grains meet
- Some gas atoms get knocked back into the grains by fission spikes: "re-solution"
- Grain face bubbles grow and can coalesce to form snake-like structures



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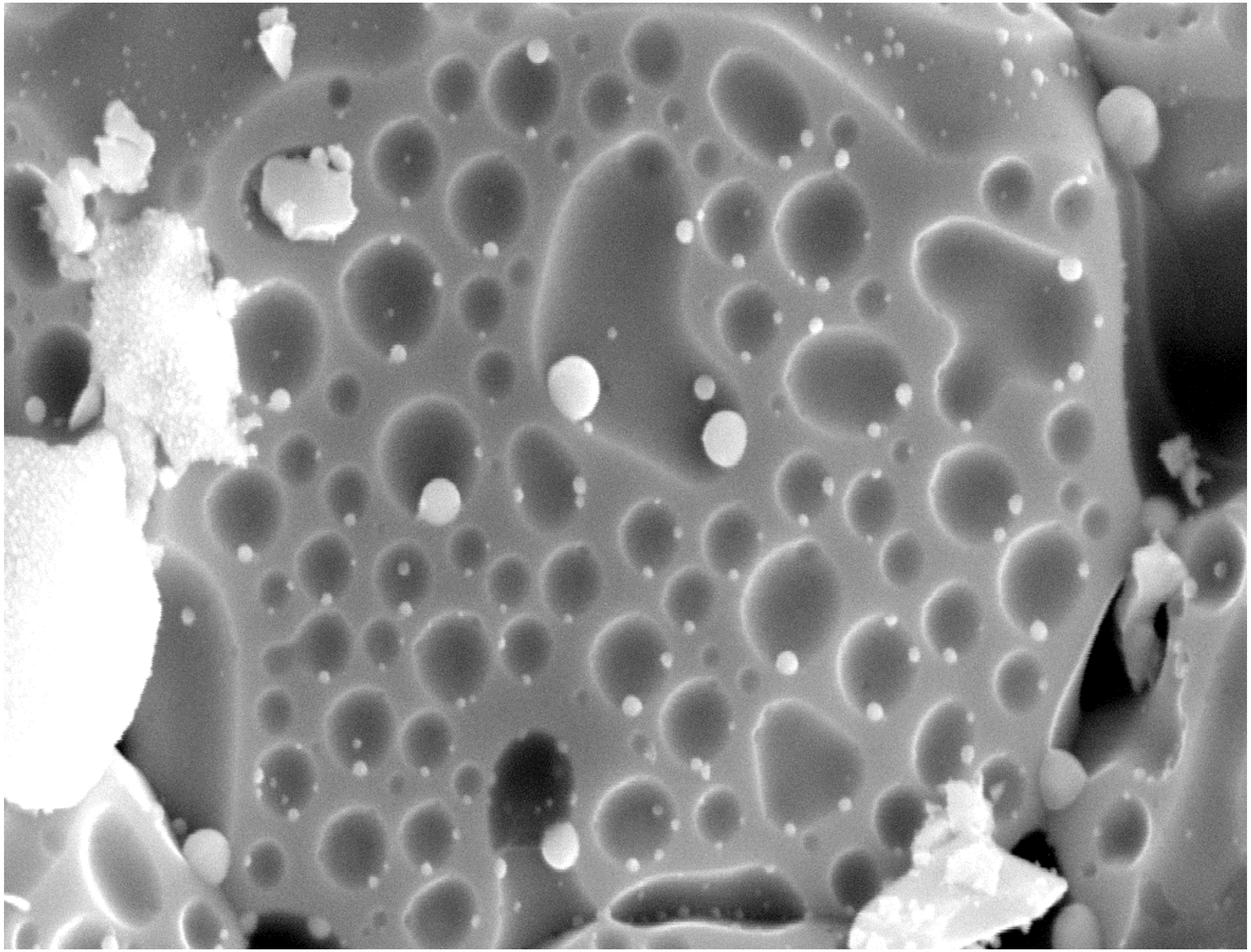
Inter-granular porosity (high burnup - coalesced pores) with metallic FP deposits



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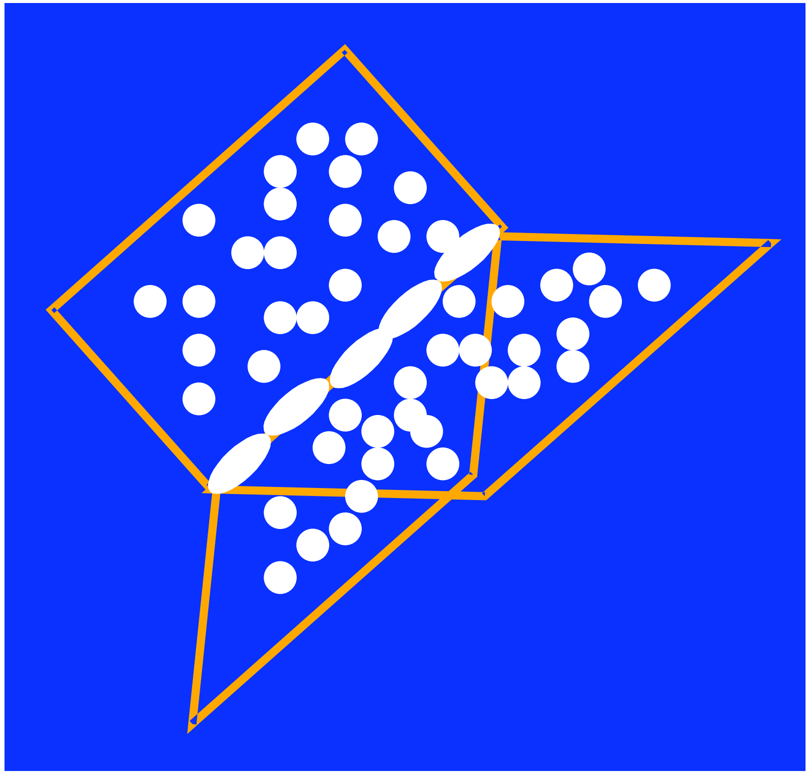
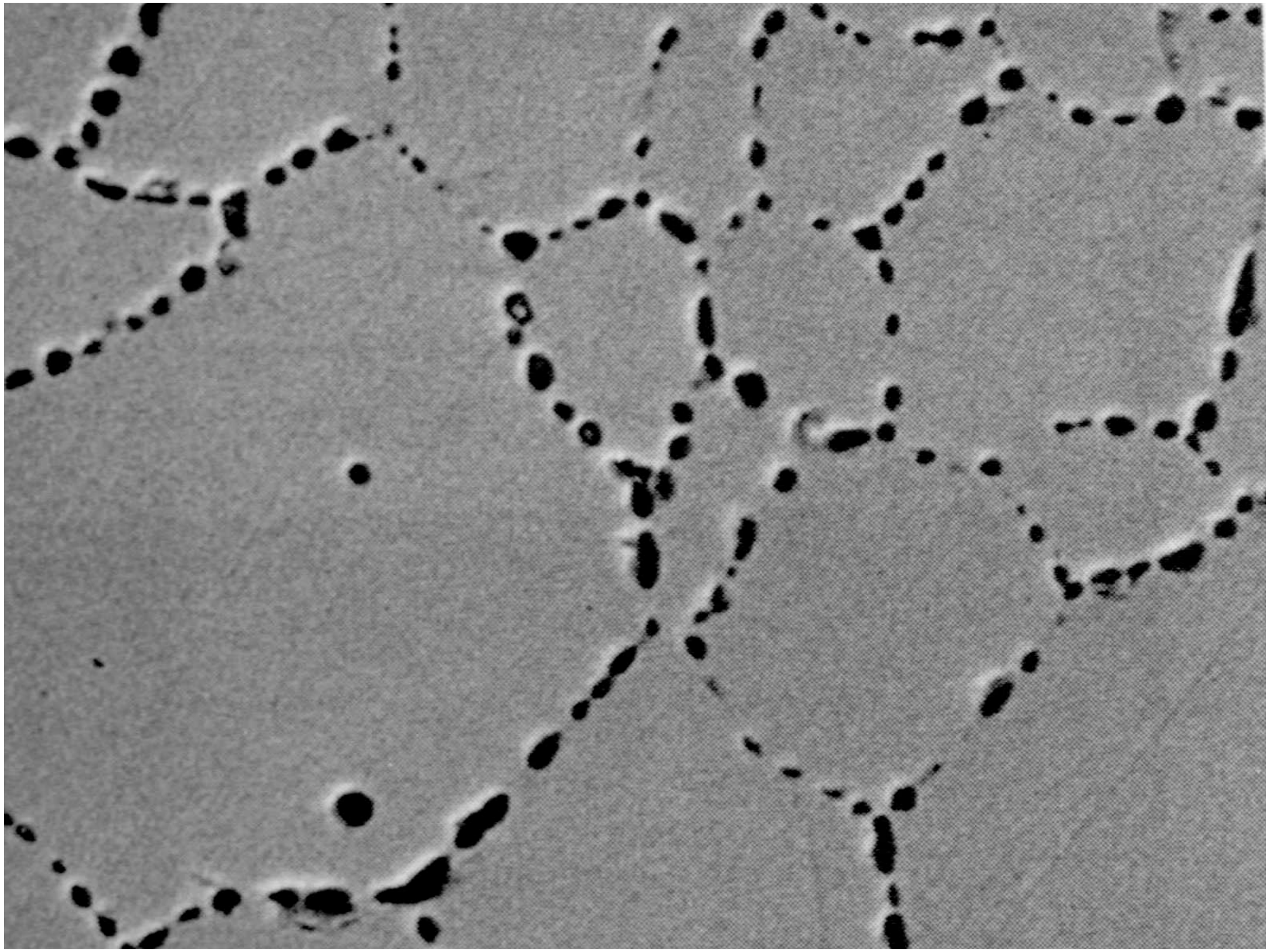
It releases fission gas

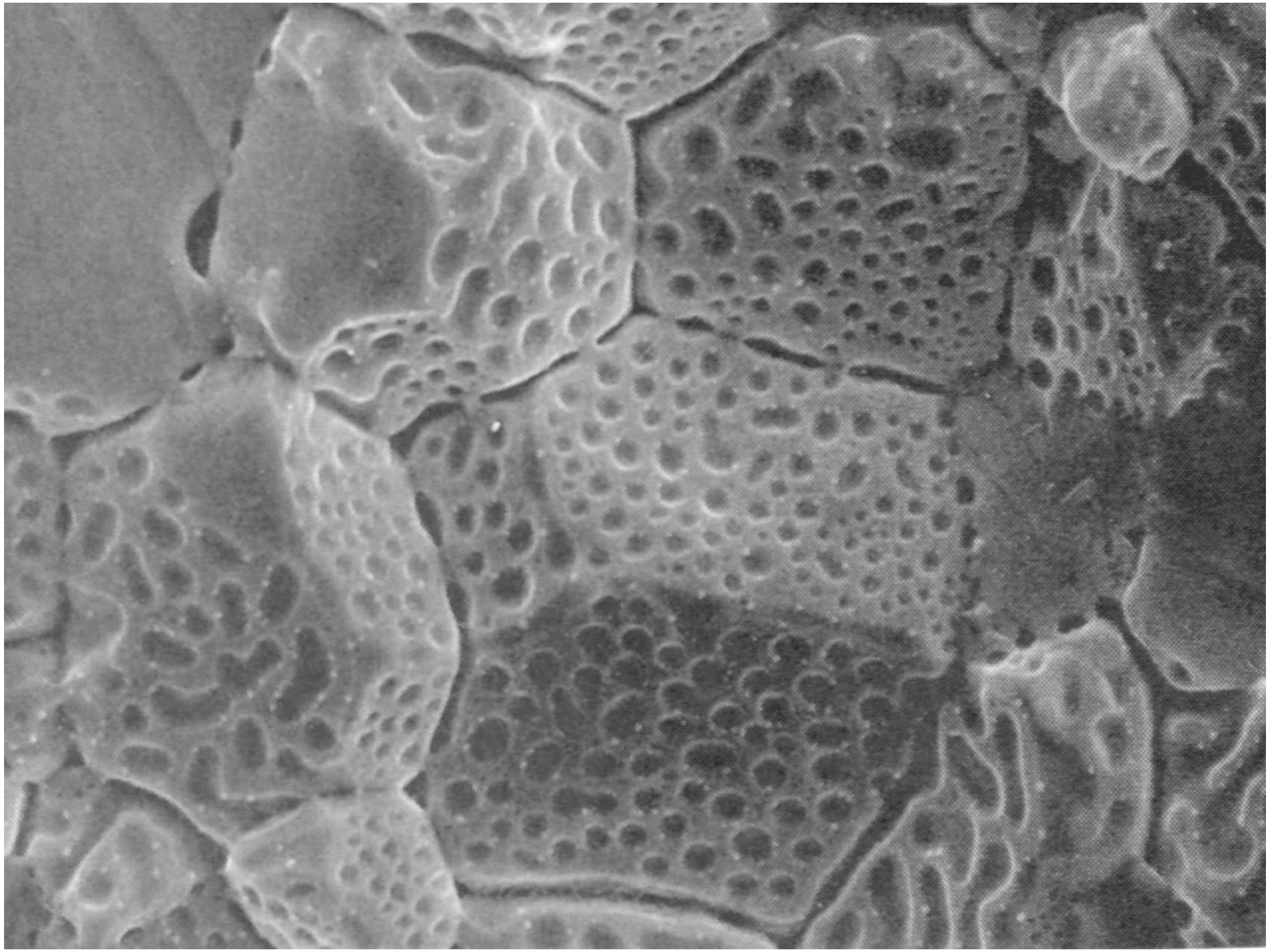


- Acicular (i.e. cigar shaped) bubbles form on the grain edges
- Grain "edges" are where 3 grains meet
- Grain edge bubbles grow and coalesce to form inter-connected pathways
  fuel is then "interlinked"
- Gas is vented to the plenum and contaminates the filling gas
- Bubbles collapse and process then repeats

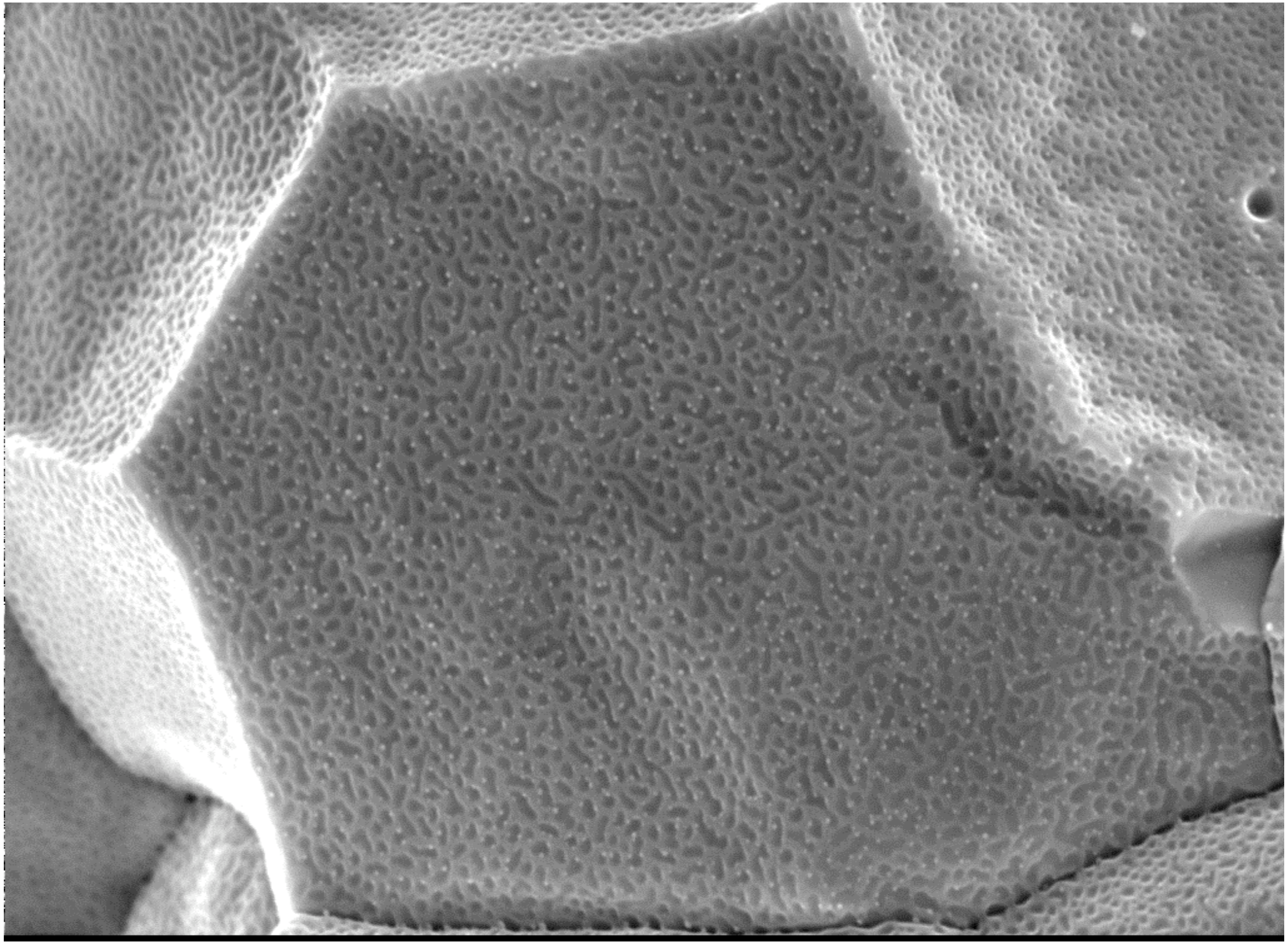
Inter-granular porosity (plan view)



Lower burnup inter-granularity



Inter-linked pores yet to develop



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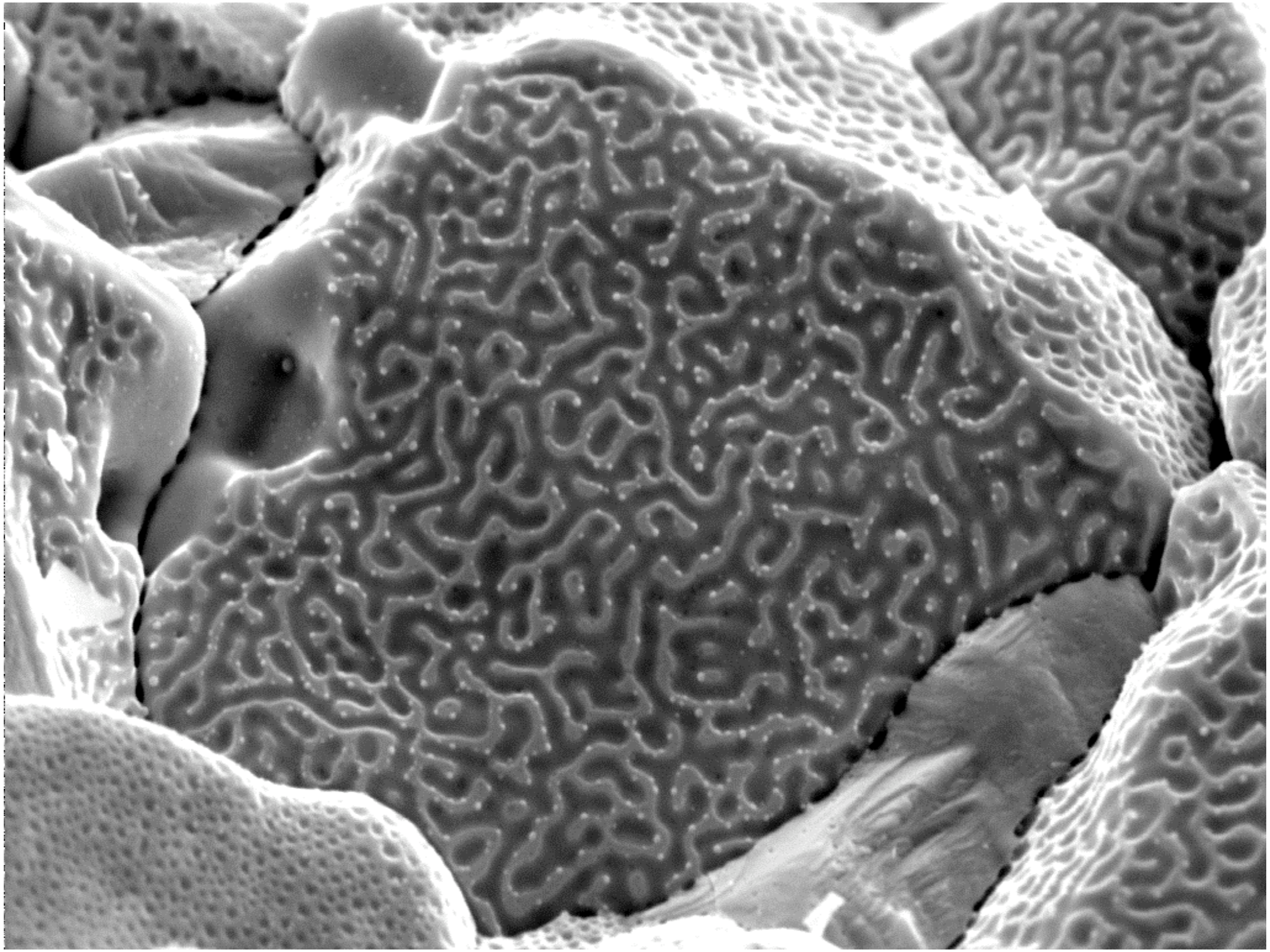
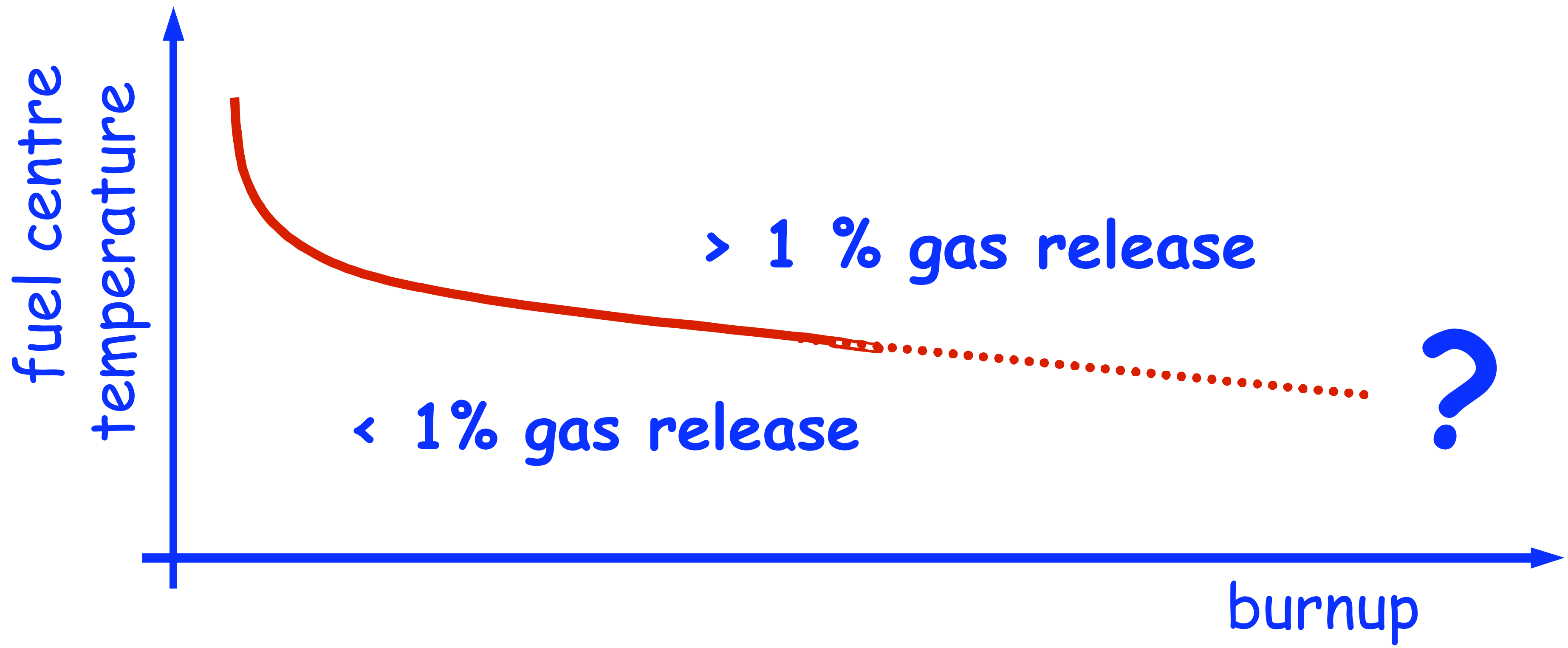
Inter-linked pores



It releases fission gas

- Empirical rule-of-thumb has been identified
- So-called “Vitanza threshold” :

$$BU^* = 0.005 \exp(9800 / Tc)$$



It gets hotter

- Fuel temperatures tend to increase at higher burnups
  - release of Xe, Kr poisons the helium filling gas
  - build-up of fission products in the fuel matrix degrades its thermal conductivity
- Higher temperatures mean more fission product
- Positive feed-back loop develops
  - gas pressure can eventually re-open the fuel-clad gap, leading to runaway thermal feed-back
  - this is one of the major life-limiting factors which constrains fuel duty

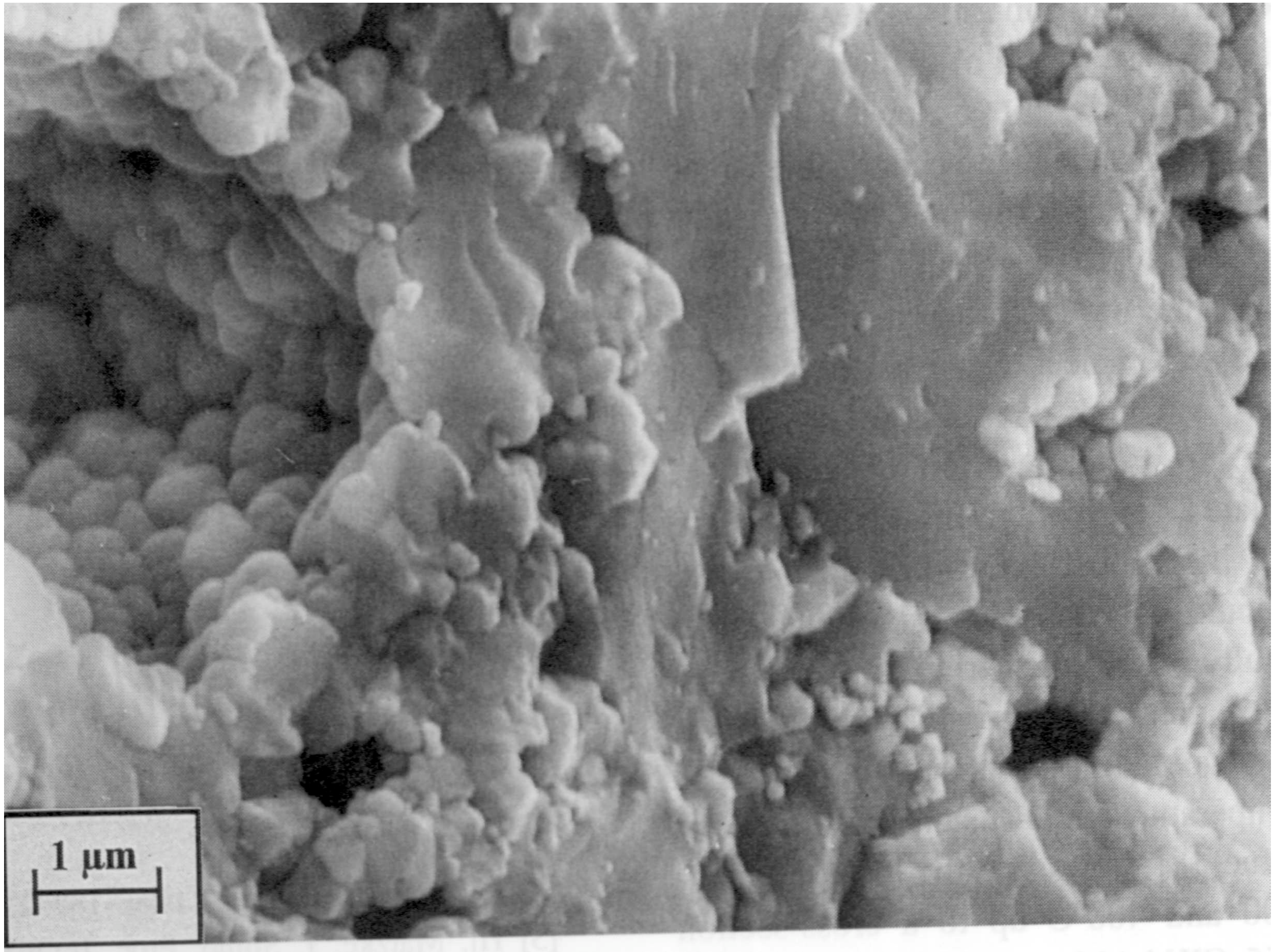
Its microstructure evolves

- By end of life, different regions of the pellet can have radically different microstructures
- As well as densification, swelling, gas release, ...
- High temperatures can lead to equi-axed grain growth
- High temperature gradients can cause porosity to migrate, giving columnar grain growth and leading to central hole formation
- High burnups lead to the “rim effect”

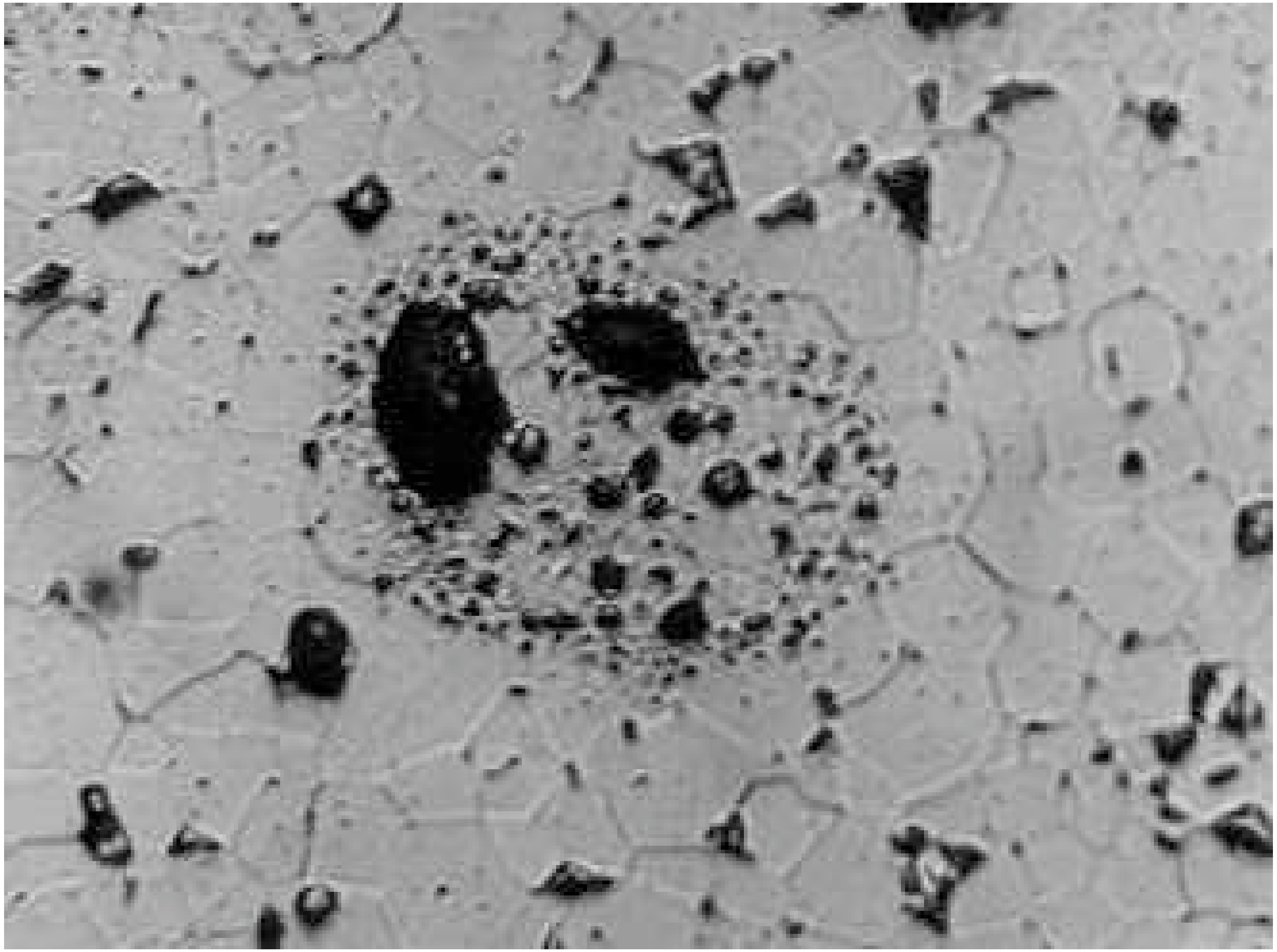
The Rim Effect

- A combination of low temperature and high fission rate
- High lattice damage, which isn't annealed out
- A new micro-structure develops
  - high density of small, Xe-filled bubbles
  - sub-micron fuel grains
- Mainly associated with outer 100-200 microns of pellet, where high Pu build-up occurs
- Also seen in Pu-rich agglomerates in MOX fuel
- At very high burnups (~70 GWd/t) can cover significant fractions of the pellet, and may lead to problems in fast transients
- Future fuels may need more resistance to formation of the "rim" structure (including near Pu-rich agglomerates)

Grain structure in rim - sub-grains inside the grain



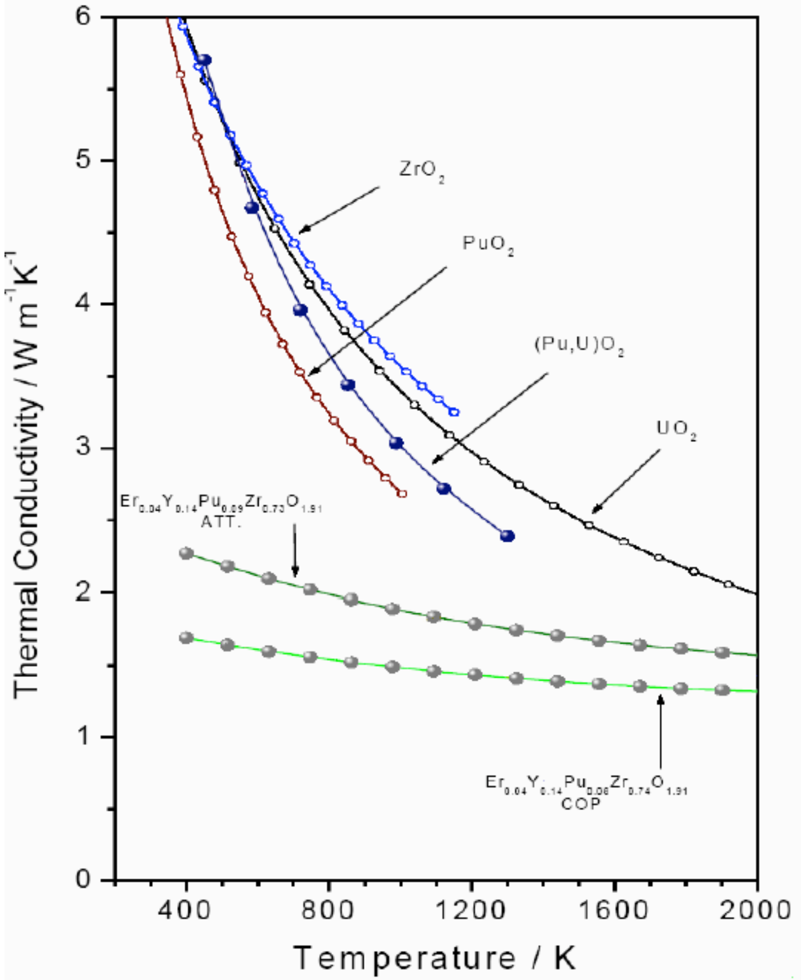
Pu rich region in MOX fuel



Sustainability

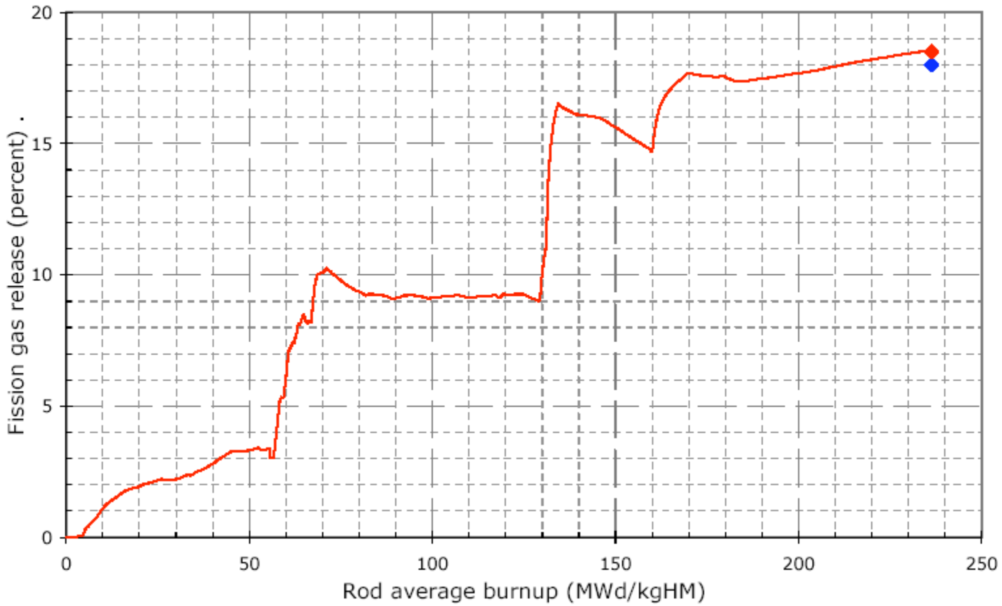
Destruction of Pu using Inert Matrix Fuels

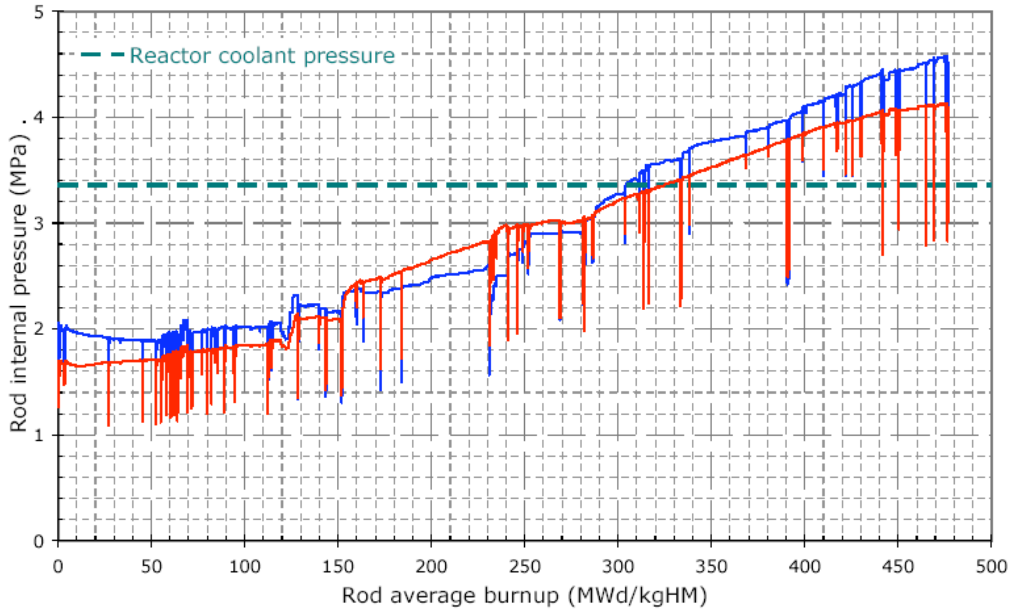
- Conventional mixed (U,Pu) oxide, MOX, contains 80-90% ^{238}U → creates new Pu by neutron capture
- Replacing U with an inert matrix would eliminate new Pu production and maximise Pu destruction rates
- Candidates include magnesia, alumina, and zirconia
- Zirconia:
  - ✓ high durability, good radiation stability, good accommodation of actinides, high melting point, small neutron cross-section, good compatibility with conventional Zr-based cladding
  - ✘ manifests different phases over temperatures of interest (but is readily stabilised using 5-10% yttria); **low thermal conductivity**
  - 😐 ability to retain fission products remains unproven; resistant to conventional reprocessing techniques



Destruction of Pu using Inert Matrix Fuels

- Analytical models for yttria-stabilised zirconia (YSZ) have been developed and validated against an experiment conducted in the Halden test reactor, using BNFL MOX fuel as a reference
- Resulting models used to calculate outcomes for YSZ fuel in commercial PWRs
- Results suggested major changes to the fuel design would be needed, and/or severe restrictions on fuel duties
- Advantages of YSZ do not merit the resulting operational and safety compromises





Summary

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Our "to do" list …

Avoid fuel failure during normal operation and frequent faults

Reduced fuel damage and degradation under accident conditions

- Understanding of failure and degradation mechanisms
- Identification and development of mitigating approaches
  - Improved cladding materials
  - Improved fuel materials

Improve fuel cycle economics

- Reduced manufacturing costs
- Simplified processes / simplified designs / reduced scrap
- Higher burnup (up to the economically optimal point)

Improve operational flexibility

- Facilitate load-follow and frequency-follow operation (tolerance to power manoeuvres)
- Longer cycle lengths
- Simplify leak detection

Improved sustainability

- Ability to burn Pu as MOX / transmutation of Pu (and Np)
- Reduced resource requirements
- Alternative fuel materials (e.g. thorium)
- Reduced environmental impact

Thanks for your attention!